STATISTICAL PHYSICS OF TWO COMPONENT DIELECTRICS

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ABSTRACT. Maximizing entropy has proven to be a very successful approach in statistical physics. Its use has led to accurate descriptions of a wide variety of physical phenomena in terms of Gibbs measures. When considering binary dielectric systems, the advantage of this approach is two fold. First, the system may be described in terms of the Hamiltonian (the energy associated with the dielectric body) and the corresponding Helmholtz potential (free energy). This analysis is facilitated by a reduced system energy representation used in homogenization theory, which allows one to express the Hamiltonian in terms of a Stieltjes transform of the measure associated with geometric resonances of the system. The description of the system using this approach parallels that of the Ising model. Numerical simulations modeling a two dimensional dielectric ElectroRheological fluid, demonstrate how the ground state of dielectric ER fluids might be predicted using this approach. Alternatively, one may use techniques of random matrix theory. The geometric resonances of the system are the eigenvalues of a random operator that lies at the very heart of two phase homogenization theory. Numerical simulations of the level spacing statistics, associated with this random operator, gives clear evidence that this system is described by a class of exactly solvable random matrix ensembles, which describe disordered mesoscopic conductors quite well. It is hoped that this approach may lead to the description of ER fluid ground states via minimization of a logarithmic potential. It is also hoped that it may characterize the conductor/insulator transition of percolating two phase conductors, as the closing of spectral gaps about the endpoints of the spectral interval.

Contents

1.	Preface	- 2
2.	Review of the Information Theoretic Approach to Statistical Mechanics	3
3.	Infinite Two Component Dielectric Systems	5
4.	Bergman–Milton Spectral Representations of the Effective Permittivity	
	for Two Component Dielectrics	6
5.	Statistical Mechanics of Binary Dielectrics and the ER Fluid	7
5.1.	Introduction	7
5.2.	Distribution and Spectral Representations of System Energies	8
5.3.	Analytic Structure of the Partition Function and Helmholtz Potential	13
5.4.	Perturbation and Asymptotic Analysis	15
5.5.	Stability Analysis, Maxwell's Relations, and Fundamental Empirical	
	Relations	17
6.	Magnetic Spin Systems–The Ising Model	19
6.1.	Abstract Lattice Systems and the Lee–Yang Theorem	20
7.	A Two Parameter Dipole Model of the ER Fluid	24

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7.1. The Model and Results	24
8. Random Matrix Theory of Disordered Systems	27
8.1. Random Matrix Theory of Mesoscopic Conductors	28
8.2. Random Matrix Theory of Two Component Conductors	31
References	

1. Preface

In the hope to make this work accessible to the mathematicians and physicists in our research group, I have tried to make it as self contained as possible. Because of the diverse backgrounds of the workers in our group, I have included details regarding the mathematics and relevant physics of the system discussed. Some sections may be review for some and necessary information for others, therefore I have included a table of contents to make navigation of this work easier. The layout is as follows.

Section 2 is a short review of the information theoretic approach to statistical mechanics. The key points are, the complete generality of the First Law of Statistical Mechanics and its differential form, under the maximum Shannon entropy Ansatz, and the connection of system energy to the Gibbs-Boltzmann distribution. Section 3 defines the infinite two component dielectric system of interest, the relevant form of Maxwell's equations, and physical quantities thereof. Section 4 briefly discusses a variational approach that leads to the orthogonal decomposition of the total electric field into its curl free and divergence free counterparts. This decomposition leads to: the resolvent representation of the electric field in terms of random operators that lie at the heart of homogenization theory, a reduced Hamiltonian which is essential to a thermodynamic description of the system, and the Bergman-Milton representation of the effective permittivity in terms of spectral measures associated with the random operators. Section 5 introduces the ER fluid and motivates the statistical mechanics approach to the problem. The introduction given in section 5.1 may be omitted on first reading. Section 5.2 goes through a detailed analysis of the general form of system energy and its decomposition into external, internal, and interactional components. Arriving at the First Law's electric work term and spectral representation of the Hamiltonian, for an infinite two component dielectric system. Sections 5.2.1, 5.2.2, and 5.2.3 discuss important special and extremal cases of the general framework and can be omitted on first reading. Section 5.3 discusses the analytic properties of the Hamiltonian, which permits a factorization of the partition function that appears to allow the Helmholtz free energy to be expressed as a logarithmic potential, relative to a measure independent of the contrast parameter. In order to understand variations in Hamiltonian and free energy, associated with changes in applied field strength and dielectric contrast, section 5.4 is devoted to a perturbation analysis. Arriving at a PDE that relates changes in free energy, corresponding to changes in applied field strength, to that of changes in dielectric contrast. An equation relating dielectric contrast derivatives of the logarithmic potential relative to the Bergman-Milton spectral measure (Golden potential) and applied field strength derivatives of the free energy, arises naturally in this discussion. At this point we have key components that permit a standard thermodynamic analysis including: consequences of Maxwell's relations, stability

 $\mathbf{2}$

analysis, and relationships between important empirical quantities. The details of these topics are given in section 5.5. The introduction given in the beginning of section 6 reviews the Ising model and may be omitted upon first reading. Section 6.1 introduces the abstract Hilbert space notation used in section 7.1 and proves the Lee-Yang theorem. This section should be omitted on first reading and used as reference. In section 7 the numerical results of a three parameter generalization of a dipole ER fluid model are discussed. The relevant details in modeling the ER fluid is discussed in the introduction of this section and can be omitted on first reading. The numerical results of this dipole model, a discussion of the associated large deviation principle, and future related numerical work is discussed in section 7.1. Numerical simulations of eigenvalue statistics, of the mentioned random operator, resemble that of a class of exactly solvable random matrix ensembles (RMEs), which describes the metal/insulator transition in MC quite well. This is introduced in the beginning of section 8 and the properties of the RME is outlined in section 8.1. The results of eigenvalue statistics, corresponding to binary materials, are given in section 8.2.

2. Review of the Information Theoretic Approach to Statistical Mechanics

In statistical physics, one is faced with the problem of assigning probabilities to events based on a few significant bits of information. In practice this information is far from sufficient to obtain objective, nor unique probabilities. It is common to use the concept of entropy in order to develop a theoretical description of the macroscopic properties, of such a system, based on its underlying microscopic properties, which are often not precisely known. Prediction of the macroscopic system behaviors, based on insufficient or incomplete data, is part of *information theory*. The ideas of information theory form a solid foundation for statistical thermophysics [26] and have been extended to systems where thermal fluctuations are not of primary importance.

An entropy function S is a measure of the amount of uncertainty in a statistical model. The idea behind entropy, is that one is not entitled to assume more knowledge, less uncertainty, than that given by subsidiary conditions, such as average values, unity measure of the probability space, *etc.* Furthermore, any assignment of probabilities that satisfy these conditions but yield a value of S other than its maximum is unjustified on the basis of known data. Therefore, the common attitude is to use probability measures which maximize the entropy, thereby maximizing the uncertainty of a system, subject to known information.

The analysis done by Shannon [1948] provides a remarkably clear quantitative measure of the uncertainty inherent in a set of probabilities [26]. In this analysis he derived the following expression widely known as the Shannon entropy

(1)
$$S(\{p_i\}) = -k \sum_i p_i \ln p_i, \quad k > 0 \text{ arbitrary.}$$

A common method for maximizing functions with given constraints is the method of Lagrange multipliers. When one only knows the average of some quantity, say $U \equiv \sum_i p_i U_i$, and that $\sum_i p_i = 1$, the resultant probability distribution is known as the canonical ensemble. It is found by maximizing $S/k - \alpha \sum_i p_i - \beta \sum_i p_i U_i$, where α and β are Lagrange multipliers. Regarding the p_i as independent variables, one arrives at $p_i = \exp(-\beta U_i)/Z$, $Z \equiv e^{\alpha+1} = \sum_i e^{-\beta U_i}$ [26]. The normalization

function Z and the probability distribution p_i of event *i* are widely known as the partition function and Gibbs-Boltzmann distribution respectively. This implies $\langle U_i \rangle = -\partial \ln Z / \partial \beta$. While this can in principle be solved for β , other procedures are used for discovering the nature of β as the solution by this method is extremely difficult [26]. What these procedures are, is of key interest for the system considered in this work. Using the definition $A \equiv -\beta^{-1} \ln Z$, widely known as the Helmholtz potential or free energy, allows one to write the entropy as [26]

(2)
$$S/k\beta = U - A = -\beta^{-1} \sum_{i} p_i [-\beta U_i - \ln Z] = \left. \frac{\partial(\beta A)}{\partial \beta} \right|_U - A.$$

Equation (2) suggests that the Helmholtz potential $A = -\beta^{-1}(\alpha + 1)$, contains the same amount of information as the entropy. Indeed we will show that every result which can be calculated from one, can be calculated from the other. Using the chain rule one can also show that the variance of U can be expressed as $\langle U_i^2 \rangle - \langle U_i \rangle^2 = \partial^2 \ln Z/\partial\beta^2$ [26].

If other subsidiary conditions are known, say the averages $\langle f_n \rangle = c_n$ of functions $f_n(U_i)$ (one always has the constraint $f_0 = 1$, $c_0 = 1$), the Gibbs-Boltzmann distribution becomes $p_i = \exp\left(-\sum_n \beta_n \langle f_n(U_i) \rangle\right)/Z$ with partition function given by $Z = \sum_i \exp\left(-\sum_n \beta_n \langle f_n(U_i) \rangle\right)$ [26]. It then follows that $\langle f_n \rangle = -\partial \ln Z/\partial \beta_n$. Equation (2) becomes $S/k = \ln Z + \sum_n \beta_n \langle f_n \rangle$ showing that S/k and $\ln Z$ are Legendre transformations of one another [26]. If we regard S as a function of the $\{\langle f_n \rangle\}$ we have $\beta_n = \partial(S/k)/\partial \langle f_n \rangle$ giving the $\{\beta_n\}$ in terms of the $\{c_n\}$. From this, we have that the symmetric matrices defined by $\partial^2(S/k)/\partial \langle f_n \rangle \partial \langle f_m \rangle = B_{n,m}$ and $-\partial^2 \ln Z/\partial \beta_r \partial \beta_j = A_{r,j}$ are inverses of one another: $A = B^{-1}$ [26].

Quantum theory identifies the $\{U_i\}$ as energy states of Hamiltonian systems and the $\{p_i\}$ as equilibrium distributions [26]. This identification has been generalized to Hamiltonian systems with a continuum of energy states, identifying the equilibrium probability (Gibbs) measure as $P(d\omega) = Z^{-1} \exp(-\beta \mathcal{H}(\omega)) d\omega$, where the space of all statistical configurations ω is denoted Ω . The macroscopic energy is given by the Hamiltonian \mathcal{H} and $Z = \int_{\Omega} P(d\omega) \exp(-\beta \mathcal{H}(\omega))$ is the corresponding partition function. To simplify notation we will continue to use that of a discrete probability space, as its generalization is clear.

The relation (2), $U = S/k\beta + A$, was obtained without making any assumptions regarding the nature of the system and is therefore a fundamental relation of statistical mechanics. It is a statement of conservation of energy and is therefore a constraint imposed on the system [4]. In order to see this, we look at the extremely useful differential form of this equation called the *First Law of Statistical Mechanics* [26]: $dU = dS/k\beta - \sum_i p_i dU_i \equiv \delta Q + \delta W$. Neither δQ nor δW are exact differentials [32][4]. The symbols δQ and δW are used to indicate the linear differential forms or pfaffins of these functions [4]. Even though neither are exact differentials their sum dU as well as dS are exact [32]. The term $\delta W = -\sum_i p_i dU_i$ (notice the minus sign) represents the differential of work done by the surroundings on the system changing the characteristic energy states $\{U_i\}$ [26]. Conservation of energy in Hamiltonian systems then identifies $-\delta W$ as the work done by the system on the surroundings [32] [9]. Various work terms may be identified by examining the physical relevance of terms, $\partial U_i/\partial z_k$, in the expansion of the differential dU_i . Where $\{z_k\}$ are the state variables which determine the eigen–energy levels U_i [26].

In the derivation of the First Law, no assumptions were made about the nature of the system or its evolutions. Therefore, it is valid for reversible, irreversible, quasi–static, and even non-quasi–static evolutions, during which the thermodynamic state cannot be defined at all [4]. For a detailed discussion of thermodynamic state, reversibility and other related concepts see [4] [32] [26].

3. Infinite Two Component Dielectric Systems

When studying the statistical mechanics of two component dielectrics, it is advantageous to use techniques of homogenization theory that lead to the effective permittivity. It is natural to consider an infinite stationary medium and work with it directly as the effective dielectric tensor, defined in section 4, is a local quantity that has nothing to do with macroscopic boundaries. The ergodicity of the framework [11] allows one to do so and puts the statistical mechanics of two component dielectrics in a natural setting.

Consider an infinite two component composite consisting of ideal (perfect electrical insulators [25]), linear, homogeneous, ideal heat conducting, incompressible, electrically neutral dielectrics of permittivity ϵ_1 and ϵ_2 , $\epsilon_2 > \epsilon_1$, at constant room temperature, with uniform density. The *closed* boundaries separating the two phases are defined by the characteristic functions χ_i of each phase. Where $\chi_1(\mathbf{x}) = 1$ for $\mathbf{x} \in S_1 \equiv {\mathbf{x} \in \mathbb{R}^d | \epsilon(\mathbf{x}) = \epsilon_1}$, the set S_2 is defined similarly, $\chi_2 = 1 - \chi_1$, and d is the dimension of the system. Define the contrast ratio $h = \epsilon_1/\epsilon_2$ and s = 1/(1-h) so that 0 < h < 1 and $1 < s < \infty$. The system is influenced by a uniform electric field \mathbf{E}_0 created by an infinite parallel plate capacitor at infinity. Initially the field strength is zero and increases quasi-statically to a nonzero value.

The equations governing the equilibrium system are that of Maxwell [16] [15]:

(3)
$$\nabla \times \mathbf{E} = 0, \quad \nabla \cdot \mathbf{D} = 0,$$
$$\epsilon_0 \nabla \cdot \mathbf{E} = \rho_t = \rho_f + \rho_b = \nabla \cdot \mathbf{D} - \nabla \cdot \mathbf{P}_0.$$

By the linearity of each material component $\mathbf{P}_0 \equiv \epsilon_0 \chi^e \mathbf{E}$ and $\mathbf{D} \equiv \epsilon \mathbf{E}$ [15]. By equations (3), Gauss's theorem, and regularity of the Lebesgue measure, for every $\mathbf{x} \in S_1 \bigcup S_2$ we have $\epsilon_0 \mathbf{E}(\mathbf{x}) = \mathbf{D}(\mathbf{x}) - \mathbf{P}_0(\mathbf{x})$. Therefore, $\epsilon = \epsilon_0(1 + \chi^e)$ where ϵ is the permittivity and χ^e is the electric susceptibility of the dielectric. Both are tensors in general but may be considered scalar functions of position by homogeneity of the individual phases. The *total* electric field is denoted \mathbf{E} while the displacement and polarization fields are denoted \mathbf{D} and \mathbf{P}_0 respectively. For slow oscillatory fields the same equations hold with ϵ considered a complex function of position [16]. The electric neutrality of the system requires that the free charge density ρ_f be identically zero, as well as the bound charge density ρ_b , on $S_1 \bigcup S_2$ as they are proportional to each other there [26]. Therefore the interior of the two component composite dielectric may be thought of as free space partitioned by a surface charge density $\sigma = \mathbf{P}_2 \cdot \mathbf{n}$, where \mathbf{n} is the outward normal to the boundaries separating the two phases [26].

Even when the applied field strength is zero, there exists a local, non-zero, macroscopic field \mathbf{E}_s due to charge distributions created by the contrast of permittivities. Linearity of equations (3) imply $\mathbf{E} = \mathbf{E}_0 + \mathbf{E}_s$. It is important to note that, although \mathbf{E}_s may be expressed as $\mathbf{E}_s = \mathbf{E} - \mathbf{E}_0$, it is actually independent of \mathbf{E}_0 . For it is, by definition, the macroscopic field present in the dielectric body in the absence of an external field. The existence and uniqueness of \mathbf{E}_s is given in section 4. The assumption of macroscopic electric neutrality requires that the volume average of \mathbf{E}_s is zero, $\langle \mathbf{E}_s \rangle_V = 0$. This suggests that this field is randomly oriented.

The astute reader may be wondering why the behavior of this infinite system should depend on the permittivity of free space ϵ_0 when the entire space is filled with materials of permittivity ϵ_1 and ϵ_2 . The reason for the appearance of ϵ_0 in equations (3) is that it was derived under the assumption that the material was localized in free space [16] [26] [15] [4]. This apparent inconsistency is easily lifted once one notices, as soon as we assume linearity of the material, we have the following identities

(4)
$$\epsilon_0 \mathbf{E} = \mathbf{D} - \mathbf{P}_0 \equiv \epsilon \mathbf{E} - (\epsilon - \epsilon_0) \mathbf{E} \iff \epsilon_2 \mathbf{E} = \mathbf{D} - \mathbf{P}_2 \equiv \epsilon \mathbf{E} - (\epsilon - \epsilon_2) \mathbf{E}.$$

Therefore ϵ_2 can be used in place of ϵ_0 without physical nor mathematical inconsistencies, given the assumptions made about the system.

4. Bergman–Milton Spectral Representations of the Effective Permittivity for Two Component Dielectrics

In this section we introduce the effective permittivity tensor and briefly discuss its origins and properties. By definition, it is the permittivity of a one component material, that stores the same amount of energy as the two component material. This definition gives a natural way of describing two component dielectrics, via statistical mechanics, as the system is modeled entirely by this energy.

Consider the Hilbert space of stationary random fields, $H \subset L^2(\Omega, \mathscr{F}, P)$, and the associated Hilbert space of stationary, curl free, random fields [11]

$$\mathscr{H} \equiv \{\mathbf{f}(\omega) \in H \mid \nabla \times \mathbf{f} = 0 \text{ weakly and } \langle \mathbf{f} \rangle_{\Omega} = 0\} \subset H,$$

where $\langle \cdot \rangle_{\Omega}$ is a suitably chosen ensemble average. Now consider the following variational problem: find $\mathbf{G} \in \mathscr{H}$ such that

(5)
$$\langle \epsilon(\mathbf{E}_0 + \mathbf{G}) \cdot \mathbf{f} \rangle_{\Omega} = 0 \quad \forall \mathbf{f} \in \mathscr{H}.$$

Under certain assumptions on $\epsilon(\mathbf{x}, \omega) \equiv \epsilon_1 \chi_1(\mathbf{x}, \omega) + \epsilon_2 \chi_2(\mathbf{x}, \omega) = \epsilon_2 (1 - \chi_1(\mathbf{x}, \omega)/s)$, this problem has the unique solution \mathbf{E}_s satisfying [11]

(6)
$$\nabla \times \mathbf{E} = 0, \quad \nabla \cdot \mathbf{D} = 0,$$

$$\mathbf{D} = \epsilon \mathbf{E}, \quad \mathbf{E} = \mathbf{E}_0 + \mathbf{E}_s, \quad \langle \mathbf{E} \rangle_{\Omega} = \mathbf{E}_0.$$

In view of (5) (6), we define the effective permittivity ϵ^* for a two component dielectric by

(7)
$$\epsilon^* \mathbf{E}_0 \equiv \langle \mathbf{D} \rangle_\Omega \iff \epsilon^* \mathbf{E}_0 \cdot \mathbf{E}_0 = \langle \mathbf{D} \cdot \mathbf{E} \rangle_\Omega$$

since equation (5) implies $\langle \mathbf{D} \cdot \mathbf{E}_s \rangle_{\Omega} = 0$ therefore $\langle \mathbf{D} \cdot \mathbf{E} \rangle_{\Omega} = \langle \mathbf{D} \cdot \mathbf{E}_0 \rangle_{\Omega}$. This can also be understood by recalling that divergence free and curl free fields are orthogonal spaces. Manipulation of (6) yields

(8)
$$\mathbf{E}_k = sR_s(\mathbf{E}_0)_k$$

where $R_s \equiv (s + \Gamma \chi_1)^{-1}$ is the resolvent of the random operator $-\Gamma \chi_1$, which is self adjoint with respect to the inner product weighted by χ_1 . The integrodifferential operator $\Gamma = -\nabla ((-\Delta)^{-1} \nabla \cdot)$ is a projection onto curl free fields and \mathbf{E}_k is the k^{th} direction of the electric field. Here we have concentrated on the diagonal

 $\mathbf{6}$

components of the symmetric tensor ϵ^* (see [11] for more details). Application of equation (8) yields

(9)
$$F(s) \equiv 1 - \epsilon^* / \epsilon_2 = \langle \chi_1 E_k / s \rangle_{\Omega} = \langle \chi_1 R_s(\mathbf{E}_0)_k \cdot (\mathbf{E}_0)_k \rangle_{\Omega}.$$

The spectral theorem then gives the Bergman–Milton representation of the effective dielectric constant [11]:

(10)
$$F(s) = \int_0^1 \frac{d\mu(\lambda)}{s-\lambda}.$$

Equation (10) represents the effective dielectric constant through a Stieltjes transformation of the positive spectral measure μ , associated with the random operator $\Gamma\chi_1$ [11]. It is well known [29] that the Bergman–Milton representation attains the following lower (upper) bounds

(11)
$$\frac{p_1}{s} = \inf_{\omega \in \Omega} F(s) \le F(s) \le \max_{\omega \in \Omega} F(s) = \frac{p_1}{s - p_2}$$

for laminates parallel (perpendicular) to the applied field respectively (see sections 5.2.2 and 5.2.3). Of course by definition, $0 \le F(s) \le 1$ for real s. This continues to hold for |F(s)| when $s \notin [0, 1]$ is considered a complex variable [11].

It is worth mentioning that, through a similar analysis, the function defined by $G(s) = 1 - \epsilon_1 (\epsilon^{-1})^* = \langle \chi_2 D_k / s \rangle$, where $(\epsilon^{-1})^* \equiv \langle (\chi_1 / \epsilon_1 + \chi_2 / \epsilon_2) D_k \rangle$, also has a representation of the form (9) (10), using $(\mathbf{D}_k)_0$ in lieu of $(\mathbf{E}_k)_0$ and interchanging (ϵ_2, χ_1) with (ϵ_1, χ_2) . Alternatively, the same formalism can be used to generate integral representations (10) for ϵ^* and $(\epsilon^{-1})^*$ using the operator $\tilde{\Gamma} = \nabla \times ((-\Delta)^{-1} \nabla \times)$ in lieu of Γ . The operator $\tilde{\Gamma}$ is a projection onto divergence free fields of Coulomb gauge. It is also worth mentioning that equations (6) govern the theory of conductors, dielectrics, magneto-statics, heat flow, and elasticity. Therefore, the effective parameter (10) is quite general. Note, throughout this paper the Euclidean norm of vectors, indicated in boldface, will be the corresponding non-bold italic symbols (e.g. $|\mathbf{E}_0| \equiv E_0$).

5. STATISTICAL MECHANICS OF BINARY DIELECTRICS AND THE ER FLUID

5.1. Introduction. It has long been speculated [13] [14] [11] [7] that the binary structure of the Ising model, or equivalently the two phase lattice gas [36] [37], may be generalized to describe the behavior of binary dielectric systems under the influence of an external electric field. Evidence that this may very well be true was indicated in a result by Golden [14]. There he showed that the Ising model two parameter critical exponent scaling relations, for the Stieltjes representation of the effective dielectric constant in the contrast variable h = 1 - 1/s and a new spectral variable $y = \lambda/(1 - \lambda)$. Thus, showing that the analytic structure of the two systems are identical. The generality of the information theoretic approach to statistical mechanics, reviewed in section 2, further indicates that the techniques of this beautiful theory may be applied to two component composites in a way that parallels the Ising model.

The purpose of this section is to further explore these connections by providing a rigorous derivation of the system Hamiltonian for a binary dielectric, in the presence of an external field. We show that the structure of the Hamiltonian and Helmholtz potential of this system and that of the Ising model are extremely similar, thus the

thermodynamic structure is also similar. We hope to extend the techniques that have been so successful in describing phase transitions in thermodynamic systems to ElectroRheological (ER) fluids.

ER fluids are examples of two component dielectrics, which exhibit an electrically induced liquid to solid phase transition. At a critical electric field strength, the solid spheres, immersed in the background fluid, form structures which increase the viscosity of the composite by several orders of magnitude, leading to a solid phase [33]. To study the phase transitions occurring in an ER fluid, consider an infinite statistically stationary emulsion of two dielectrics subject to an external electric field \mathbf{E}_0 . The continuous phase a dielectric liquid of permittivity ϵ_1 , while the dispersed phase consists of dielectric suspensions of permittivity ϵ_2 , with fixed geometry such as spheres. The force on each particle in the suspension is due to the multipole-multipole interactions induced by the applied field and dielectric contrast between the solid particles and liquid. When the inclusions are impenetrable spheres, increasing the applied field strength causes the particles to aggregate, forming chains then body centered tetragonal columns, along the applied field direction [33]. The formation of columns cause the ER fluid to exhibit increased viscosity or even solid-like behavior, able to sustain shear in the direction perpendicular to the applied electric field. The sharp increase in viscosity to a glass like solid happens in a few milliseconds after the external field surpasses a critical strength E_c . The rheological variation is reversed when the field strength is reduced below E_c [33]. If sensors are used to trigger the applied electric field, one can turn many devices such as clutches, valves, dampers etc. into active mechanical elements capable of responding to environmental variations [33].

5.2. Distribution and Spectral Representations of System Energies. When modeling a system using statistical mechanics, it is very important to understand the various associated energies. As the system is modeled entirely by these energetic contributions. There are many different kinds of energy associated with the infinite polarized binary dielectric being considered. First, there is the energy associated with the macroscopic field \mathbf{E}_s of the dielectric body in the absence of the applied field \mathbf{E}_0 . Second, there is the energy associated with the transducer in the absence of the two component medium, i.e. in free space. Finally, there is the energy associated with the interaction of the external field and the dielectric body. A further decomposition could identify "material energies" like $\epsilon_2 \langle \chi_1 \mathbf{E} \cdot \mathbf{E}_0 \rangle_V$ where the geometry, given by χ_1 , appears explicitly in the volume average $\langle \cdot \rangle_V$. We could also identify "pure field energies" like $\epsilon_2 \langle \mathbf{E} \cdot \mathbf{E}_0 \rangle_V$ where geometric contributions are given implicitly in the total electric field \mathbf{E} . We will see that the framework used in homogenization theory gives a detailed account of all such energies in terms of the Bergman-Milton spectral measure.

The key to a useful representation of the system Hamiltonian is the resolvent representation of the electric field found by Golden (8) [11]: $\mathbf{E} \equiv E_0 s R_s \mathbf{e}_0$, where $\mathbf{e}_0 \equiv \mathbf{E}_0/E_0$. The analysis done in section 4 and the ergodicity of the system [11] yields the following important relations:

(12)
$$\mathbf{E}_0 = \langle \mathbf{E} \rangle_\Omega = \langle \mathbf{E} \rangle_V \equiv \lim_{V \to \infty} \frac{1}{|V|} \int_V \mathbf{E}(\mathbf{x}) d^d x,$$

where $\langle \cdot \rangle_{\Omega}$ denotes an appropriate ensemble average and $V \subset \mathbb{R}^d$. An important consequence of the variational formulation of equations (6) is the following well

known result [11]. The minimum of the variational form (5), hence $\langle \frac{1}{2} \mathbf{D} \cdot \mathbf{E} \rangle_V$, satisfies $\nabla \cdot (\epsilon \nabla \phi) = 0$ weakly. Therefore, we will use this as the system energy per-unit-volume (puv) [16]. A beautiful consequence the electric field's resolvent representation is that it decouples the geometric information of the system, contained in the operator $\Gamma \chi_1$, from the dielectric contrast parameter *s* and the applied field strength E_0 . This, and equation (7), allows one to write the system energy puv in an especially simple form:

(13)
$$\left\langle \frac{1}{2} \mathbf{D} \cdot \mathbf{E} \right\rangle_V = \left\langle \frac{1}{2} \mathbf{D} \cdot \mathbf{E}_0 \right\rangle_V = \frac{1}{2} \epsilon_2 E_0^2 (1 - F(s, \omega)) = \frac{1}{2} \epsilon_2 E_0^2 \epsilon^*(s, \omega),$$

weakly for every $\omega \in \Omega$. Here $F(s, \omega) = \langle \chi_1 R_s \mathbf{e}_0 \cdot \mathbf{e}_0 \rangle_V$ has integral representation (10), whereby use of the volume average and the explicit dependence on the random operator χ_1 , the spectral measure $d\mu(\lambda, \omega)$ now depends explicitly on $\omega \in \Omega$.

Using the functional form of the spectral theorem [24] we have, for any Borel measurable function G, $\langle \chi_1 G(\Gamma \chi_1) \mathbf{e_0} \cdot \mathbf{e_0} \rangle_V = \int_0^1 G(\lambda) d\mu(\lambda, \omega)$. Using this and $\langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = 0$, one can find Stieltjes integral representations for all material and pure field energies, e.g.

(14)
$$\langle \chi_2 E_s^2 \rangle_V / E_0^2 = \langle \chi_2 \| (sR_s - I)\mathbf{e}_0 \|^2 \rangle_V = \frac{p_2}{s} + \int_0^1 \frac{\lambda(1-\lambda)d\mu(\lambda,\omega)}{(s-\lambda)^2}.$$

Integral representations for all other variants, (e.g. $\langle \epsilon_2 E^2 \rangle_V$), can also be derived. The consequences of this detailed information on the partitions of energy is unclear, but it should facilitate a detailed description and concise representation of what is considered work, internal energy, etc.

In this spirit, we follow the suggestive analysis done by Robertson on homogeneous dielectrics [26]. First, write the energy puv as

(15)
$$\left\langle \frac{1}{2} \epsilon \left(E_0^2 + 2\mathbf{E}_s \cdot \mathbf{E}_0 + E_s^2 \right) \right\rangle_V \equiv \mathcal{W}_0 + \mathcal{W}_{int} + \mathcal{W}_s = \mathcal{W}_0 + \frac{1}{2} \mathcal{W}_{int},$$

since $\langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = 0$ implies $\frac{1}{2} \mathcal{W}_{int} + \mathcal{W}_s = 0$. Using the resolvent representation of the electric field (8), recalling $\langle \mathbf{E}_s \rangle_V = \langle \mathbf{E} - \mathbf{E}_0 \rangle_V = 0$ and $\epsilon = \epsilon_2 (1 - \chi_1 / s)$, we find

(16)
$$\mathcal{W}_0 + \frac{1}{2}\mathcal{W}_{int} = \frac{1}{2}\epsilon_2 E_0^2 \left(1 - \frac{p_1}{s} - \left(F(s,\omega) - \frac{p_1}{s}\right)\right).$$

In order to make natural connections to physics, make the following definitions

(17)
$$\chi^*(s,\omega) = F(s,\omega) - \frac{p_1}{s}, \quad \chi_{\parallel}(s) = \frac{p_1}{s},$$
$$P^*(s,\omega) = \epsilon_2 \chi^*(s,\omega) E_0, \quad P^*_{\parallel}(s) = \epsilon_2 \chi^*_{\parallel}(s) E_0$$

where $\chi^* \geq 0$ denotes the effective permeability and P^* the effective polarization of the binary dielectric. Using equations (17), equation (15) may be physically summarized by

(18)
$$\mathcal{W}_{0} + \frac{1}{2}\mathcal{W}_{int} = \frac{1}{2}\epsilon_{2}E_{0}^{2}\left(1 + \chi_{\parallel}^{*}(s) - \chi^{*}(s,\omega)\right)$$
$$= \frac{1}{2}E_{0}\left(\epsilon_{2}E_{0} + P_{\parallel}^{*}(s) - P^{*}(s,\omega)\right)$$

Each term in equations (15) must be analyzed in order to correctly obtain the electric work term, to be inserted in the First Law for binary dielectrics, and Hamiltonian. By the linearity of Maxwell's equations, the Hamiltonian is the sum of

Coulomb potential energy terms, representing the potential energy of the charged particles within the system, in the fields of all charged particles both within the system and in the surroundings. The mutual energies of the particles within the system are macroscopically regarded as part of the internal energy [26]. More specifically, for a zero external field strength there still exists a nonzero microscopic field energy density. The question is, how is this energy accounted for in the macroscopic continuum description? This energy cannot be viewed as a mere shift in the zero level of internal energy because it is dependent on inter-particle distances and is therefore, in general, density and temperature dependent. Thus, this energy effectively contributes to internal energy changes and must be included in the thermodynamic internal energy of the system [4]. The mutual energies of the external particles are not part of the Hamiltonian as these interactions are no more of interest than that of the heat bath in thermal systems. Therefore, when considering the electric work done on a binary dielectric, the important energies to understand are the mutual energies of the external/internal interactions. Where do these external/internal energies belong? The surprising answer is that the microscopic Hamiltonian places the entire energy within the system! Therefore the macroscopic treatment must also do so in order to be compatible [26].

The term W_0 represents the mutual energy puv of the external charges in the presence of a two component dielectric composite of laminates parallel to the applied field. The analysis done in section 5.2.2 shows that this interaction is independent of the surface charge distribution within the dielectric. Therefore, this term is not included in the Hamiltonian. The term W_s , analogous to the self interaction term of the Ising model, is the self–energy puv of the dielectric in its state of polarization, independent of the external charges. Therefore it is regarded as part of the internal energy and is included in the system Hamiltonian. The term W_{int} represents the external charges interacting with and polarizing a homogeneous dielectric with susceptibility $\chi^*(s, \omega)$. One may be inclined to partition this energy between the the system and surroundings but the previous discussion already indicated that it must be regarded as part of the system energy [26].

Therefore, the term that is to be inserted into the First Law for binary dielectrics is, $\langle \mathcal{H}_{int} \rangle_{\Omega} \equiv \langle -\mathcal{W}_{int} \rangle_{\Omega} \equiv P^*(s)E_0$, which has Stieltjes integral representation

(19)
$$\langle \mathcal{H}_{int} \rangle_{\Omega} = \epsilon_2 E_0^2 \left\langle \int_0^1 \frac{d\mu(\lambda,\omega)}{s-\lambda} - \frac{p_1}{s} \right\rangle_{\Omega} = \epsilon_2 E_0^2 \left(\int_0^1 \frac{\langle d\mu(\lambda,\omega) \rangle_{\Omega}}{s-\lambda} - \frac{p_1}{s} \right)$$

by a Fubini theorem, where $\langle \cdot \rangle_{\Omega}$ denotes the Gibbs ensemble average. The Hamiltonian puv in the absence of externally produced electric fields is given by $\langle \mathcal{H}_s \rangle_{\Omega} \equiv \langle -\mathcal{W}_s \rangle_{\Omega}$, which has a Stieltjes integral representation that can be found using equation (14) and its variants. If one did so, after a little algebra one would find, as expected, $\mathcal{H}_s(\omega) = -\mathcal{H}_{int}/2$. Thus, the Hamiltonian puv of the system is

(20)
$$\mathcal{H}(\omega) = \mathcal{H}_{int}(\omega) + \mathcal{H}_s(\omega) = \frac{1}{2}\epsilon_2 E_0^2 \left(F(s,\omega) - \frac{p_1}{s}\right) = \frac{1}{2}P^*(s,\omega)E_0$$

This analysis demonstrates the competition between the work done on the system and geometric affects. This form of the Hamiltonian makes physical sense as we want to maximize ϵ^* [33], or equivalently minimize $F(s,\omega)$; minimizing $F(s,\omega)$ increases the Gibbs factor $\exp(-\beta\epsilon_2 E_0^2 F(s,\omega)/2)$.

The appearance of E_0 in Stieltjes integral representations of self energies (14) is due to the use of linearity of Maxwell's equations. Again, by definition, these

energies are independent of the applied field. Remarkably, the orthogonality of divergence free and curl free fields, leading to $\langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = 0$, requires that *exactly* half of the external/internal interaction energy is counteracted by the internal interactions of the system, independent of both the external field strength and configuration $\omega \in \Omega$. This suggests that the resultant charge density that forms on the contrast boundaries, as a consequence of the applied field, does so in a way that conserves the equality $\mathcal{W}_s + \frac{1}{2}\mathcal{W}_{int} = 0$ for every configuration $\omega \in \Omega$ and external field strength, weakly. This integral constraint seems to impose some sort of energy conservation on the system. This conservation is mathematically summarized by the integral equation (14) and its variants.

The following sections give a detailed analysis of some important extremal cases of the above framework. Extremal cases are central to the theory of two component composites as they are the building blocks of all composites [21]. It is also illustrative to see how the reduced Hamiltonian follows directly from boundary conditions and $\langle \mathbf{E} \rangle_V = \mathbf{E}_0$ in these cases. In these sections we will use the notation $\langle \chi_i \rangle_V = p_i$ for the volume fraction of material component *i*. Not to be confused with the probability of event *i* use elsewhere in the text.

5.2.1. Homogeneous system. For a homogeneous system $\epsilon \equiv \epsilon_2$. Therefore equation (12) implies the system energy puv is given by $\mathcal{H} = (1/2)\epsilon_2 E_0^2$, which is the interaction energy puv of the external charges in the presence of an infinite dielectric of uniform permittivity ϵ_2 . This this can be understood by noticing ϵ_2 contains no geometric information and it is assumed ρ_f and ρ_b are identically zero on the interior of S_2 (see the introduction to section 3). Therefore, the interaction of an infinite ideal dielectric of permittivity ϵ_2 , is the same as interaction with free space, other than the use of ϵ_0 . As expected, equation (12) indicates $\mathcal{W}_{int} = 0 = \mathcal{W}_s$ which makes sense as there are no internal/internal nor external/internal interactions associated with empty space. It is important to note that this follows directly from equation (6) and thus requires that we neglect the interaction with the boundary at infinity. This practice is equivalent to placing the energy of the surface integral terms, which are usually thrown away in the derivation of electric energy densities [15], as a part of the internal energy.

5.2.2. Laminates Parallel to the Applied Field. This two component system, shown in figure 1, is known [29] to have effective permittivity $\epsilon^* = p_1\epsilon_1 + p_2\epsilon_2$ and Bergman–Milton representation $F(s) = p_1/s$ (see section 4). The total electric field being curl free (6) causes the tangential component of the electric field to be continuous [16], $(\mathbf{E}_i - \mathbf{E}_{i+1}) \times \mathbf{n} = 0$, where **n** is the unit normal to the contrast boundaries. This and symmetry implies $\mathbf{E}_i = \mathbf{E}_{i+1}$ for all *i*, therefore $\mathbf{E}_i = \mathbf{E}_j$ for all *i*, *j*. Thus, set $\mathbf{E}_1 \equiv \mathbf{E}(\mathbf{x})$ for $\mathbf{x} \in {\mathbf{x} | \epsilon(\mathbf{x}) = \epsilon_1} \equiv S_1$. Equation (12) then implies

$$\mathbf{E}_0 = \langle \mathbf{E} \rangle_V = \sum_i p_i \mathbf{E}_i = \mathbf{E}_1 \sum_i p_i = \mathbf{E}_1.$$

Therefore, there is no surface charge distribution between the different regions induced by \mathbf{E}_0 [16]. The total field being identified with the applied field along with equation (8) implies $R_s = I/s$, therefore $\Gamma \chi_1 = 0$.

The following result is not limited to two component materials. Therefore using the general definitions $\epsilon = \sum_{i} \epsilon_{i} \chi_{i}$, $\epsilon_{\parallel}^{*} = \sum_{i} \epsilon_{i} p_{i}$, and that $\mathbf{E} \equiv \mathbf{E}_{0}$ yields the

system energy puv:

(21)
$$\left\langle \frac{1}{2} \mathbf{D} \cdot \mathbf{E} \right\rangle_{V} \equiv \left\langle \frac{1}{2} \left(\sum_{i} \epsilon_{i} \chi_{i} \right) E_{0}^{2} \right\rangle_{V} = \frac{1}{2} \epsilon_{\parallel}^{*} E_{0}^{2} = \frac{1}{2} \epsilon_{2} E_{0}^{2} \left(1 - \frac{p_{1}}{s} \right),$$

in accordance with the general theory, where the last equality is true for two component materials. Equation (15) implies $\mathcal{W}_s = \mathcal{W}_{int} = 0$ therefore the Hamiltonian is identically zero. Thus, the Helmholtz potential can be considered constant.





(b) Laminates perpendicular to the applied field

FIGURE 1. Extremal Geometries of the Effective Permittivity

5.2.3. Laminates Perpendicular to the Applied Field. For the two component system of laminates perpendicular to the applied field, shown in figure 1, it is known that the effective permittivity is $\epsilon^* = 1/(p_1/\epsilon_1 + p_2/\epsilon_2)$ with Bergman–Milton representation $F(s) = p_1/(s - p_2)$ [29] (see section 4). It turns out the results here, as with parallel laminates, are not limited to two component materials.

The Displacement field being divergence free (6) causes the normal component of the displacement field to be continuous: $(\epsilon_i \mathbf{E}_i - \epsilon_{i+1} \mathbf{E}_{i+1}) \cdot \mathbf{n} = 0$ for all i, where **n** is the unit normal to the boundaries [16]. This and symmetry implies $\epsilon_i \mathbf{E}_i = \epsilon_{i+1} \mathbf{E}_{i+1}$ for all i, thus $\epsilon_i \mathbf{E}_i = \epsilon_j \mathbf{E}_j$ for all i, j. Therefore, set $\mathbf{E}_1 \equiv \mathbf{E}(\mathbf{x})$ for $\mathbf{x} \in {\mathbf{x} | \epsilon(\mathbf{x}) = \epsilon_1} \equiv S_1$ then $\mathbf{E}_i = (\epsilon_1/\epsilon_i)\mathbf{E}_1$ for all i. Equation (12) then yields

(22)
$$\mathbf{E}_0 = \langle \mathbf{E} \rangle_V = \sum_i p_i \mathbf{E}_i = \epsilon_1 \mathbf{E}_1 \sum_i \frac{p_i}{\epsilon_i} \equiv \frac{\epsilon_1}{\epsilon_\perp^*} \mathbf{E}_1$$

so that $\mathbf{E}_i = (\epsilon_{\perp}^* / \epsilon_i) \mathbf{E}_0$. This imposes a global continuity equation for the displacement field:

(23)
$$(\mathbf{D} - \mathbf{D}_0) \cdot \mathbf{n} \equiv \epsilon_i \chi_i E_i - \epsilon_{\perp}^* E_0 = 0.$$

Thus, the system energy puv is

(24)
$$\left\langle \frac{1}{2} \mathbf{D} \cdot \mathbf{E} \right\rangle_{V} \equiv \frac{1}{2} \sum_{i} \epsilon_{i} p_{i} E_{i}^{2} = \frac{1}{2} \sum_{i} \epsilon_{i} p_{i} \left((\epsilon_{\perp}^{*} / \epsilon_{i}) E_{0} \right)^{2} = \frac{1}{2} \epsilon_{\perp}^{*} E_{0}^{2}.$$

in accordance with the general theory.

It is illustrative to see how the boundary condition and the orthonormality of the χ_i , $\chi_i \chi_j = \chi_i \delta_{ij}$, gives

$$\langle \mathbf{E} - \mathbf{E}_0 \rangle_V = 0 \iff \epsilon_i \chi_i E_i - \epsilon_{\perp}^* E_0 = 0 \iff \langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = 0.$$

and the partition of system energy. Indeed, if equation (23) holds then $\langle \mathbf{E}_s \rangle_V = 0$ as can easily be seen by using the orthonormality of the $\chi_i, \chi_i \chi_j = \chi_i \delta_{ij}$, to write

$$\langle \mathbf{E}_s \rangle_V = \langle \mathbf{E} - \mathbf{E}_0 \rangle_V = \left\langle \sum_i \chi_i (\mathbf{E}_i - \mathbf{E}_0) \right\rangle_V = \mathbf{E}_0 \sum_i (\epsilon_\perp^* / \epsilon_i - 1) p_i = 0.$$

The orthornormality $\chi_i \chi_j = \delta_{ij} \chi_i$ and equation (23) yields

(25)
$$\langle \epsilon E_s^2 \rangle_V = \left\langle \left(\sum_i \epsilon_i \chi_i \right) \left(\sum_i \chi_i \left(\frac{\epsilon_{\perp}^*}{\epsilon_i} - 1 \right)^2 \right) E_0^2 \right\rangle_V = (\epsilon_{\parallel}^* - \epsilon_{\perp}^*) E_0^2$$
$$\langle \epsilon \mathbf{E}_s \cdot \mathbf{E}_0 \rangle_V = E_0 \left\langle \sum_i \chi_i (\epsilon_{\perp}^* - \epsilon_i) E_0 \right\rangle_V = (\epsilon_{\perp}^* - \epsilon_{\parallel}^*) E_0^2$$
$$\langle \epsilon E_0^2 \rangle_V = \left\langle \sum_i \epsilon_i \chi_i E_0^2 \right\rangle_V = \epsilon_{\parallel}^* E_0^2$$

so that $\langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = 0$. Conversely,

$$0 = \langle \mathbf{D} \cdot \mathbf{E}_s \rangle_V = \left\langle \sum_i \chi_i \epsilon_i (E_0 (E_i - E_0) + (E_i - E_0)^2) \right\rangle_V = \epsilon_1 E_1 \left(\frac{\epsilon_1}{\epsilon_\perp^*} E_1 - E_0 \right)$$

which, with the boundary condition $E_i = (\epsilon_1/\epsilon_i)E_1$, yields equation (23). It is worth noting, as in the general setting (14), all variants of material and pure field energies (25) may be calculated for the case of laminates perpendicular to the field.

By the discussion in section 5 and equation (25), the system Hamiltonian puv is given by

$$-\mathcal{H} = \mathcal{W}_{int} + \mathcal{W}_s = \langle \epsilon \mathbf{E}_s \cdot \mathbf{E}_0 \rangle_V + \frac{1}{2} \langle \epsilon E_s^2 \rangle_V = \frac{1}{2} (\epsilon_{\perp}^* - \epsilon_{\parallel}^*) E_0^2 = -\frac{1}{2} P^*(s) E_0.$$

in accordance to the general theory, where $P^*(s) = \epsilon_2(p_1/(s-p_2)-p_1/s)E_0$ for two component materials. The configuration independence of the Hamiltonian and the E_0 independence of \mathcal{W}_s implies.

(26)
$$A = -\beta^{-1}|\Omega| + \frac{1}{2}P^*(s)E_0, \quad \frac{\partial A}{\partial E_0} = P^*(s), \quad \frac{\partial^2 A}{\partial E_0^2} = \epsilon_2 \chi^*(s) \ge 0.$$

There seems to be a sign problem here. We know that the ER fluid maximizes ϵ^* [33] so we want the Gibbs factor to be proportional to $\exp(-\alpha F(s,\omega))$ but stability requires $\partial^2 A/\partial E_0^2 \leq 0$ (see section 5.5). This seems to be contradictory. The negative sign introduced in the free energy in ([33] equation 2) would alleviate this but I don't understand where this comes from.

5.3. Analytic Structure of the Partition Function and Helmholtz Potential. In this section we explore the analytic structure of the partition function and Helmholtz potential. It is our hope to uncover properties thereof that parallel the Lee-Yang structure of the Ising model Helmholtz potential (see section 6.1) [9] [27] [36] [37]. Ultimately, we would like to obtain, from first principles in physics, a logarithmic potential representation of the Helmholtz potential. Whereby the employment of variational principles, its minimization may determine the ground states of ER fluids. The analysis done in the introduction of section 5 demonstrated that the partition function of a two component composites is

(27)
$$Z = \sum_{\omega \in \Omega} \exp\left(\alpha(p_1/s - F(s,\omega))\right) \equiv z^{p_1/s} \sum_{\omega \in \Omega} z^{-F(s,\omega)}$$

where $z \equiv \exp(\alpha)$ and $\alpha \equiv \beta \epsilon_2 E_0^2/2$. The configuration independence of p_1/s allows us to define a statistically equivalent partition function $\tilde{Z} \equiv Z/z^{p_1/s}$ with Helmholtz potential puv

(28)
$$A = -\beta^{-1} \ln \left(\sum_{\omega \in \Omega} z^{-F(s,\omega)} \right)$$

Recall [10] [2] that for finite, $|\Omega| < \infty$, random capacitor networks

(29)
$$F_n(s,\omega) = \int_0^1 \frac{d\mu_n(\lambda,\omega)}{s-\lambda} = \sum_{i=1}^n \frac{\mu_i}{s-\lambda_i} = \frac{Q_n(s,\omega)}{P_n(s,\omega)}$$

where $\{P_n(s,\omega)\}, \{Q_n(s,\omega)\}\$ are the polynomials, of order n and n-1 respectively, orthogonal to the measure $\mu_n(\lambda,\omega)$. Furthermore, the zeros of $\{P_n(s,\omega)\}\$ and $\{Q_n(s,\omega)\}\$ are precisely the poles and zeros of $F_n(s,\omega)$, in the interval [0,1], respectively. Therefore, for s outside this interval, which we will assume throughout this section, the Hamiltonian is an analytic function of its parameters [11]. In the limit $n \to \infty$, these sets of polynomials are complete in $L^2(\mu,\omega)$, for each $\omega \in \Omega$ [2]. We have $\exp(-\alpha F_n(s,\omega)) \in L^2(\mu,\omega)$ for all n by, the triangle inequality for Radon-Stieltjes integrals [31], the positivity of $\mu_n(\omega)$, and that $\int_0^1 d\mu(\lambda) = p_1$, the volume fraction of type 1 capacitors. Therefore, starting with a large finite system, represent each of the finite number $|\Omega|$ of terms, in equation (28) as a finite, truncated, sum in the denominator basis up to order M(n) < n, $\lim_{n \to \infty} M(n) = \infty$:

$$(30) Z = \sum_{\omega=1}^{|\Omega|} \exp\left(-\alpha F_n(s,\omega)\right) = \sum_{\omega=1}^{|\Omega|} \left(\sum_{j=1}^{M(n)} a_{j,n} P_j(s,\omega) + O\left(M(n)^{-\delta_{n,\omega}}\right)\right)$$
$$\equiv \prod_{i=1}^{M(n)} (s-s_i) + O\left(M(n)^{-\delta_n}\right),$$
$$a_{j,n} = \int_0^1 P_j(\lambda',\omega) \exp\left(-\alpha F_n(\lambda',\omega)\right) d\mu(\lambda',\omega),$$

for some $\delta_n = \inf_{\omega} \delta_{n,\omega} > 0$, which exists by the completeness of $L^2(\mu, \omega)$ and in general depends on s. Thus, it appears the Weierstrass theorem allows one to write the Helmholtz potential, up to statistical equivalence, as

$$A\beta = -\ln\left(\prod_{i=1}^{M(n)} (s - s_i) + O\left(M(n)^{-\delta_n}\right)\right) = -\sum_{i=1}^{M(n)} \ln(s - s_i) + O\left(M(n)^{-\eta}\right)$$

for some $\eta > 0$. The $\{s_i\}$ are determined by the $\{a_{j,n}\}$ which depend only on $\alpha = \beta \epsilon_2 E_0^2/2$ and the geometric resonances of the system and are independent of contrast parameter s. Thus, up to statistical equivalence,

(32)
$$A\beta \to -\int_0^1 \ln(s-\lambda)d\nu(\lambda) \text{ as } n \to \infty$$

14

(31)

where ν is the contrast independent measure corresponding to the distribution of the $\{s_i\}$ as $n \to \infty$.

In the canonical ensemble, it is not the average total energy that is minimized at equilibrium, but rather the Helmholtz potential [6]. Therefore, if the above analysis can be put in a rigorous mathematical language, equilibrium is synonymous with minimization of the above logarithmic potential over measures ν . It is well known [2] that the empirical measures associated with the zeros of these orthogonal polynomials minimize logarithmic potentials over $supp(\mu_n(\lambda))$. Thus, it would be natural that the measure $\nu(\lambda)$ also does, subject to certain constraints. The relationship between $\nu(\lambda)$ and $\mu(\lambda)$ is a central question.

It is worth mentioning that for non-oscillatory fields, each term in the partition function is real and positive. Therefore, away from a phase transition, the real zeros thereof are in one-to-one correspondence with the geometric resonances. Whether or not these constitute all roots of the partition function is a central question. Furthermore, as $\alpha \to \infty$ the $a_{j,n} \to 0$ for all j, n, by the positivity of $F(s, \omega)$. This would cause a singularity in the Helmholtz potential which would indicate an electrically induced phase transition.

5.4. Perturbation and Asymptotic Analysis. In section 5.3 it was demonstrated how the statistically equivalent Helmholtz potential (28) might be expressed as a logarithmic potential with respect to a contrast independent measure $\nu(\lambda)$. By construction, this measure is determined by $\alpha = \beta \epsilon_2 E_0/2$ and the geometric resonances of the system. Hence the measures $\mu(\lambda, \omega)$, each of which are determined by configurational, $\omega \in \Omega$, geometric resonances. The measure $\mu(\lambda) = \langle \mu(\lambda, \omega) \rangle_{\Omega}$ is determined by geometric resonances, and by α and s through the use of the Gibbs ensemble average $\langle \cdot \rangle_{\Omega}$. Using this logarithmic representation and the statistically equivalent Helmholtz potential (28) we get the following formal relationships between the measures ν and μ .

$$(33) \quad A = -\beta^{-1} \int_{supp(\nu)} \ln(s-\lambda) d\nu[\alpha](\lambda) = -\beta^{-1} \ln \sum_{\omega \in \Omega} \exp\left(-\frac{\beta}{2}\epsilon_2 E_0^2 F(s,\omega)\right)$$
$$\frac{\partial A}{\partial s} = -\beta^{-1} \int_{supp(\nu)} \frac{d\nu[\alpha](\lambda)}{s-\lambda} = -\frac{1}{2}\epsilon_2 E_0^2 \int_0^1 \frac{\langle d\mu(\lambda,\omega) \rangle_{\Omega}}{(s-\lambda)^2}$$
$$\frac{\partial A}{\partial E_0} = -\beta^{-1} \int_{supp(\nu)} \ln(s-\lambda) d\left[\frac{\partial\nu[\alpha]}{\partial E_0}\right](\lambda) = \epsilon_2 E_0 \int_0^1 \frac{\langle d\mu(\lambda,\omega) \rangle_{\Omega}}{s-\lambda}$$

where $supp(\mu) \subseteq [0, 1]$. In order to hopefully shed some light on these relationships, this section is devoted to a perturbation and asymptotic analysis of the system. In this section we will always use the statistically equivalent Hamiltonian and associated partition function and Helmholtz potential (28).

It is important to note that for $s \notin [0, 1]$, which we will assume throughout this section, the Hamiltonian is an analytic function of its parameters [11]. Therefore, when a parameter in the Hamiltonian is perturbed, $\xi \mapsto \xi + \delta \xi$, $\delta \xi \ll 1$, the affect on the system Hamiltonian can be represented generally as $\mathcal{H}(\omega) \mapsto \mathcal{H}(\omega) + \delta \mathcal{H}(\omega)$, $\delta \mathcal{H}(\omega) \ll 1$, given the uniform boundedness of all parameters. Denoting the Gibbs probability distribution and its perturbation by $P(d\omega)$ and $\delta P(d\omega)$ respectively one finds [5]

(34)
$$\delta P(d\omega) = P(d\omega)(1 - \beta (\delta \mathcal{H}(\omega) - \langle \delta \mathcal{H}(\omega) \rangle_{\Omega}) + O[(\beta \delta \mathcal{H}(\omega))^{2}])$$
$$\delta Z = Z(1 - \beta \langle \delta \mathcal{H} \rangle_{\Omega} + O[\langle (\beta \delta \mathcal{H}(\omega))^{2} \rangle_{\Omega}])$$
$$\delta A = A + \langle \delta \mathcal{H} \rangle_{\Omega} + O[\langle \beta (\delta \mathcal{H}(\omega))^{2} \rangle_{\Omega}]).$$

The Hamiltonian of the binary dielectric is (20) $\mathcal{H} = -\epsilon_2 E_0^2 F(s,\omega)/2$. Therefore

(35)
$$\delta_s \mathcal{H}(\omega) = -\frac{1}{2} \epsilon_2 E_0^2 \frac{\partial F(s,\omega)}{\partial s} \delta s + O[(\delta s)^2]$$
$$\delta_{E_0} \mathcal{H}(\omega) = -\epsilon_2 E_0 F(s,\omega) \delta E_0 + O[(\delta E_0)^2].$$

It is important to note that the especially simple representation of the system Hamiltonian, with respect to its parameters, will facilitate a perturbation analysis of all orders. For brevity we only consider the first order expansion here.

A question of interest is: given δE_0 what δs results in the same change in system energy $\delta \mathcal{H}(\omega)$, to first order. By equation (35) and the positivity of $F(s, \omega)$ for all $\omega \in \Omega$, the answer is given by the following relation

(36)
$$\frac{\delta E_0}{\delta s} = \frac{E_0}{2} \frac{\partial}{\partial s} \ln F(s,\omega)$$

One could also ask: given δE_0 what δs results in the same average change in system energy to first order (equivalently Helmholtz potential or partition function by equation (34)). By equations (34) (35) (43) and the positivity of $F(s, \omega)$ one can show that the following statements are equivalent

(37)
$$\frac{\delta E_0}{\delta s} = \frac{E_0}{2F(s)} \left\langle \frac{\partial F(s,\omega)}{\partial s} \right\rangle_{\Omega} \equiv \gamma(s, E_0)$$
$$\left\langle \delta_s \mathcal{H}(\omega) - \delta_{E_0} \mathcal{H}(\omega) \right\rangle_{\Omega} = O[(\delta E_0)^2 + (\delta s)^2]$$
$$\frac{\partial A}{\partial s} \delta s - \frac{\partial A}{\partial E_0} \delta E_0 = O[\beta \left((\delta E_0)^2 + (\delta s)^2 \right)]$$
$$\delta_s A - \delta_{E_0} A = O[\beta \left((\delta E_0)^2 + (\delta s)^2 \right)],$$

where $F(s) \equiv \langle F(s,\omega) \rangle_{\Omega}$. For notational simplicity denote partial derivatives in s by a prime, e.g. $F'(s) = \partial F(s)/\partial s$. The following property of the Gibbs-Boltzmann distribution

(38)
$$P'(d\omega) \equiv (\exp(-\alpha F(s,\omega))/Z)' = -\alpha (F'(s,\omega) - \langle F'(s,\omega) \rangle_{\Omega}) P(d\omega)$$

yields a commutator

(39)
$$\langle F(s,\omega)\rangle'_{\Omega} - \langle F'(s,\omega)\rangle_{\Omega} = -\alpha \text{Cov}(F(s,\omega),F'(s,\omega))$$

allowing equation (37) to be written, up to first order, as

$$\frac{\delta A}{\delta s} = \gamma(s, E_0) \frac{\delta A}{\delta E_0}, \quad \gamma(s, E_0) = \frac{E_0}{2} (\ln F(s))' - \frac{E_0 \alpha}{2F(s)} \operatorname{Cov}(F(s, \omega), F'(s, \omega)).$$

By equations (34) (35), as $\delta E_0, \delta s \to 0$ the variational derivatives of the Helmholtz potential converge to the partial derivatives. If $\delta E_0, \delta s \to 0$ in a way that preserves

the relationships of equations (37). In this limit, if it exists, equation (40) defines a PDE relating the change of Helmholtz potential in s to that in E_0 :

(41)
$$\frac{\partial A}{\partial s} = \gamma(s, E_0) \frac{\partial A}{\partial E_0}$$

Equation (39) shows that as "thermal" energy dominates $(\alpha \ll 1)$ long rang correlations are not present, the commutator is zero, and the *s* dependence of $\langle \mu(\lambda, \omega) \rangle_{\Omega}$ is negligible. Although, for $\alpha \gg 1$ correlations become significant suggesting that there is long range correlations in the material.

We conclude this section with a brief discussion to hopefully illuminate a connection between the response function F(s) and a corresponding logarithmic potential. To do so, make the definitions $\phi(s,\omega) = \int_0^1 \ln(s-\lambda)d\mu(\lambda,\omega), \ \phi(s) \equiv \langle \phi(s,\omega) \rangle_{\Omega} = \int_0^1 \ln(s-\lambda)\langle d\mu(\lambda,\omega) \rangle_{\Omega}$ so that $F(s,\omega) = \phi'(s,\omega)$. Repeated use of equation (39) yields

$$\phi'(s) - \frac{1}{\epsilon_2 E_0} \frac{\partial A}{\partial E_0} = \phi'(s) - F(s) = -\alpha \left(\langle \phi(s,\omega) F'(s,\omega) \rangle_\Omega - \phi(s) F'(s) \right) + \alpha^2 \phi(s) \operatorname{Cov} \left(F(s,\omega), F'(s,\omega) \right).$$

If some of these terms can be shown to be small and/or approximately independent, this may give us $\phi'(s) - \frac{1}{\epsilon_2 E_0} (\partial A / \partial E_0) \approx 0$ which would support that the Helmholtz potential may have a logarithmic potential representation with respect to the Bergman-Milton spectral measure.

5.5. Stability Analysis, Maxwell's Relations, and Fundamental Empirical Relations. As mentioned before, the key to an explicit statistical mechanics model of binary dielectrics, is the explicit decoupling of the applied field strength E_0 and contrast parameter *s* from complicated geometric affects, provided by the resolvent representation of the electric field (8). This permits the following standard "thermodynamic" analysis of the system. For notational simplicity we make the following definitions $P^* = \langle P^*(s,\omega) \rangle_{\Omega}, \ \chi^* = \langle \chi^*(s,\omega) \rangle_{\Omega}, \ F(s) = \langle F(s,\omega) \rangle_{\Omega}, \ T = (k\beta)^{-1}, C_r = T(\partial S/\partial T)_r, \ \alpha_r = (\partial P^*/\partial T)_r, \ \text{and} \ \chi^e_r = (\partial P^*/\partial E_0)_r/\epsilon_2$ where *T* is the "temperature," C_r is the specific heat at constant $r, \ \alpha_r$ is a quantity analogous to the volume expansivity, χ^e_S is the isentropic susceptibility, χ^e_T is the isothermal susceptibility, and $\langle \cdot \rangle_{\Omega}$ denotes the Gibbs ensemble average. The quantities α_r , χ^e_S , and χ^e_T are examples of response functions which are extremely important in thermodynamics due to their use in the fluctuation dissipation theorem [5].

In section 2 it was argued that the First Law $dU = dS/k\beta - \sum_i p_i dU_i$ is a fundamental relation of statistical mechanics; we will denote the probability space as discrete for notational simplicity. It was also mentioned that various work terms can be identified by expanding the exact differential dU_i in terms of its parameters. From the analysis done in section 5.2 we see that the electric energy of binary dielectric systems depends only on two parameters: the external field E_0 and the dielectric contrast s. Expanding dU_i in these variables we have

(42)
$$dU_i = \frac{\partial U_i}{\partial E_0} \Big|_s dE_0 + \frac{\partial U_i}{\partial s} \Big|_{E_0} ds.$$

The first term $(\partial U_i/\partial E_0)_s$ has units of polarization. It can be argued since the average $\langle (\partial U_i/\partial E_0)_s dE_0 \rangle_\Omega$ can be understood only as the work done by the external field to change the energy levels of the system, thus polarizing the dielectric

body, this term must be the polarization. The analysis done in section 5.2 thus identifies this term as P^*dE_0 . The second term has units of energy. We will denote the average of this term by ψ . The Helmholtz potential, and various other thermodynamic potentials are obtained via Legendre transformations of the internal energy: $A(T, E_0, s) = U(T, E_0, s) - TS(T, E_0, s)$, etc. [4]. Since all thermodynamic potentials can be obtained from one another [9] we focus on the Helmholtz and Gibbs potential, A and G respectively. The following equation gives the differential forms of these equations:

(43)
$$dA(T, E_0, s) = -S(T, E_0, s)dT - P^*(T, E_0, s)dE_0 - \psi(T, E_0, s)ds, dG(T, P^*, s) = -S(T, P^*, s)dT + E_0(T, P^*, s)dP^* - \psi(T, E_0, s)ds.$$

The functions of state are obtained by obvious differentiations of thermodynamic potentials (e.g. $(\partial A/\partial E_0)_{T,s} = -P^*(T, E_0, s) \leq 0$). Maxwell's relations are obtained under the assumption that derivatives of thermodynamic potentials commute, e.g.

(44)
$$-\frac{\partial^2 A}{\partial E_0 \partial s}\Big|_T = \frac{\partial P^*}{\partial s}\Big|_T = \frac{\partial \psi}{\partial E_0}\Big|_T$$
$$\frac{\partial^2 G}{\partial P^* \partial s}\Big|_T = \frac{\partial E_0}{\partial s}\Big|_T = -\frac{\partial \psi}{\partial P^*}\Big|_T.$$

By equations (17) and (44) we have

(45)
$$\frac{\partial P^*}{\partial s}\Big|_T = -\epsilon_2 E_0 \left(\int_0^1 \frac{\langle d\mu(\lambda,\mu) \rangle_\Omega}{(s-\lambda)^2} - \frac{p_1}{s^2} \right)$$

so that

(46)
$$\psi = -\left.\frac{\partial A}{\partial s}\right|_T = -\frac{1}{2}\epsilon_2 E_0^2 \left(\int_0^1 \frac{\langle d\mu(\lambda,\mu)\rangle_{\Omega}}{(s-\lambda)^2} - \frac{p_1}{s^2}\right)$$

plus some function g(T, s) which is zero since for $E_0 = 0$ the system is assumed electrically neutral, macroscopically, which suggests that for $E_0 \ll 1$ we have $\psi = \langle (\partial U_i/\partial s)_{E_0,T} \ll 1$ thus $g(T, s) \ll 1$ which implies g(T, s) = 0 as it is independent of E_0 . Equations (44) (17), and properties of partial differentiation, also gives

(47)
$$\frac{\partial E_0}{\partial s}\Big|_T = -\frac{\partial \psi}{\partial E_0}\Big|_T \left(\frac{\partial P^*}{\partial E_0}\right)_T^{-1} = -\frac{\partial P^*}{\partial s}\Big|_T \left(\frac{\partial P^*}{\partial E_0}\right)_T^{-1} = -E_0\frac{\partial}{\partial s}\ln(\chi^*(s)),$$

where we have used $P^*(s) = \epsilon_2 \chi^*(s) E_0$, making connection to equation (40) since:

(48)
$$\frac{\partial A}{\partial s}\Big|_{T} = \frac{\partial A}{\partial E_{0}}\Big|_{T} \frac{\partial E_{0}}{\partial s}\Big|_{T} = -E_{0}\frac{\partial}{\partial s}\ln(\chi^{*}(s))\frac{\partial A}{\partial E_{0}}\Big|_{T}$$

Thermodynamic equilibrium is a state of minima of thermodynamic potentials, subject to appropriate constraints. Its origin may be understood through the Le Chatelier Principle, which may be stated quite generally as follows [26]: Any local departures from equilibrium (fluctuations) that arise in a stable system induce changes in the intensive parameters that diminish these fluctuations. Equilibrium of fluctuating systems can also be understood through The Fluctuation Dissipation Theorem [5]. The general result is that thermodynamic potentials are convex functions of their extensive variables and concave functions of their intensive variables.

These conditions, restricted by a differential-based analysis, apply only to the local shape of the potential surfaces [26]. Thus, for stability, it is necessary that the Helmholtz potential be, locally, a concave function of its intensive parameters and a convex function of its extensive parameters. This means that [26] [9]

(49)
$$\frac{\partial^2 A}{\partial T^2}\Big|_{E_0,s} = -\frac{\partial S}{\partial T}\Big|_{E_0,s} = -\frac{C_{P^*}}{T} \le 0$$

or $C_{P^*} \geq 0$. Since $P^* = \epsilon_2 E_0 \chi^*$,

(50)
$$\frac{\partial^2 A}{\partial E_0^2}\Big|_{T,s} = -\frac{\partial P^*}{\partial E_0}\Big|_{T,s} = -\epsilon_2 \chi^* = -\epsilon_2 \left(F(s) - \frac{p_1}{s}\right) \le 0$$

which, by equation (11), is always true. Equation (50) simply states that the effective polarization should increase as the applied field strength increases. This equation also yields the identification $\chi_T^e = \chi^*$ showing that F(s) is a response function and therefore has a connection to the fluctuation dissipation theorem [5]. By equation (46) we have

(51)
$$\frac{\partial^2 A}{\partial s^2}\Big|_{T,E_0} = -\frac{\partial \psi}{\partial s}\Big|_{T,E_0} = -\frac{1}{2}\epsilon_2 E_0^2 \left(\int_0^1 \frac{\langle d\mu(\lambda,\mu)\rangle_{\Omega}}{(s-\lambda)^3} - \frac{p_1}{s^3}\right) \le 0$$

or $\int_0^1 \langle d\mu(\lambda,\mu) \rangle_{\Omega}/(s-\lambda)^3 \ge p_1/s^3$. By the definitions made in the introduction of this section and the rules of partial differentiation, we have the following relations between empirical parameters and response functions, for fixed contrast s [26]:

(52)
$$C_{E_0} - C_{P^*} = \frac{T\alpha_{E_0}^2}{\epsilon_2 \chi_T^e} = \epsilon_2 T \chi_T^e \alpha_{P^*}^2$$
$$\chi_T^e - \chi_S^e = \frac{\alpha_{E_0}^2 T}{\epsilon_2 C_{E_0}}$$
$$\frac{\chi_S^e}{\chi_T^e} = \frac{C_{P^*}}{C_{E_0}}.$$

Relationships of empirical functions involving the contrast parameter will be explored in a later version of this paper.

6. Magnetic Spin Systems-The Ising Model

For magnetic systems the microscopic objects are spins, rather than particles. This stems from the fact that magnetic monopoles do not exist [15]. The simplest possible model of a spin system is the Ising model. A general Ising lattice is a regular array of elements, each of which can interact with other elements in the lattice and with an external field. The non-kinetic part of the Hamiltonian is [26]

(53)
$$\mathcal{H} = -\sum_{i} V_i(\sigma_i) - \frac{1}{2} \sum_{i,j}' U_{i,j}(\sigma_i, \sigma_j).$$

The symbol \sum' denotes the omission of the self energy (i = j terms), the 1/2factor allows for both $U_{i,j} = U_{j,i}$ terms in the sum, and $V_i(\sigma_i)$ is a non-interaction term that may represent the effect of an external field or its equivalent [26]. As a simplification, the spins may be thought of as vectors occupying fixed points in space.

Consider a system $\Lambda \subset \mathbb{Z}^d$ that consists of N such spins $\{\sigma_i\}_{i=1:N}$, where the spin variables are rescaled so that $\|\sigma_i\| = 1$, in the presence of a uniform external magnetic field H. The classical Ising model simplifies things even further by requiring $\sigma_i \in \{-1, 1\}$. The Hamiltonian and partition function for the resultant configuration space, $\Omega = \{-1, 1\}^N$, are

(54)
$$\mathcal{H}_{\Lambda}(H,\beta) = -H \sum_{i \in \Lambda} \sigma_i - \frac{1}{2} \sum_{i,j \in \Lambda}' J_{ij} \sigma_i \sigma_j$$
$$Z_{\Lambda}(H,\beta) = C_N \int_{\omega \in \Omega} d\omega \exp\left(-\beta \mathcal{H}_{\Lambda}\right)$$

where where the constant C_N is chosen to make the thermodynamics extensive [32], $m(\Lambda) = \sum_{i \in \Lambda} \sigma_i$ is the magnetization of the configuration, and $J_{ij} = J$ for the i, jnearest neighbors, zero otherwise. The interaction strength J > 0 for ferromagnets and J < 0 for antiferromagnets. The magnetization is found through the Helmholtz potential: $m(\Lambda) = -\partial A/\partial H$, $A = -\beta^{-1} \ln Z$.

In 1952 T. D. Lee and C. N. Yang showed that the root distribution of the grand partition function completely determines the equation of state in statistical phase transition theory [36]. In particular, the properties of the system, in relation to phase transitions, are determined by the behavior of the roots near the positive real axis. Furthermore, the problem of the ferromagnetic Ising model in a magnetic field is mathematically identical with that of a "lattice gas" [37]. It was also shown that the equation of state of the condensed phases as well as the gas phase can be correctly obtained from a knowledge of the distribution of roots [36] [37]. Section 6.1 places the Ising model in the language of abstract Hilbert space and proves the Lee–Yang theorem. It is within this abstract setting that we will study the phase transitions of the dipole ER fluid discussed in section 7.

6.1. Abstract Lattice Systems and the Lee–Yang Theorem. The following construction of lattice systems and the proof of the Lee–Yang theorem is due to Ruelle [27]. Consider a *d* dimensional lattice $\Lambda \subset \mathbb{Z}^d$. Associated with a lattice site $x \in \Lambda$ is an occupation number, or equivalently a lattice site state number, $n_x =$ $0, \ldots, N$. Let there be *V* sites and consider the set of all possible configurations of the system within $\Lambda \equiv \{x_i\}_{i=1}^V, x_i \in \mathbb{Z}^d$. There are $(N+1)^V$ such configurations parameterized by the *V*-tuples $(n_{x_1}, \ldots, n_{x_V})$ where $\sum_{i=1}^V n_{x_i} = n$. The set of "admissible functions" $\{f_\alpha\}$ on $x \in \Lambda$, \mathscr{H}_x , defines a Hilbert space [27] of dimension N+1. A natural definition of the Hilbert space on Λ is given by $\mathscr{H}_\Lambda \equiv \bigotimes_{x \in \Lambda} \mathscr{H}_x$ of dimension $(N+1)^V$. Each $f_\alpha \in \mathscr{H}_\Lambda$ is a self adjoint operator, bounded below, with spectrum consisting of isolated eigen–values of finite multiplicity [27].

Typically a potential energy acts on the particles which is independent of momentum. In this case the Hamiltonian is $\mathcal{H}_{\Lambda} = \sum_{i=1}^{n} p_i^2/2m + U_{\Lambda}(x_1, \ldots, x_n)$, where p_i and m, are the momentum and mass of particle *i* respectively. In systems such as this, the statistical weight due to kinetics trivially cancels in all calculations [32][27]. Therefore, we may make the identification $\mathcal{H}_{\Lambda} \equiv U_{\Lambda}$ where U_{Λ} summarizes all interactions of the system with the environment and within the system itself.

To further develop the binary structure of the classical ferromagnetic Ising model, consider a lattice gas with site state number N = 1 so that each site $x \in \Lambda$ is in state zero or one, e.g. corresponding to spin down and spin up respectively. The canonical ensemble is a measure, $Z_{\Lambda}^{-1} \exp(-\beta \mathcal{H}_{\Lambda}) d\tau$, on the set of all possible configurations of the system within Λ , where τ is the measure corresponding to the Bernoulli random variable on Λ . Since the mutual site interaction locations are uniquely determined by the boundary of the system $\partial \Lambda$ and the state one particles, the potential energy may be written [27]

(55)
$$U_{\Phi}(X) \equiv U_{\Lambda}(X) \equiv \sum_{Y \subset X} \Phi(Y), \quad X \equiv \{x \in \Lambda | n_x = 1\}.$$

Theorem 6.1. [27] Define $\|\Phi\| \equiv \sum_{0 \in X} |\Phi(X)| / N(X)$ where N(X) is the cardinality of X.

(i) The set ℬ of potentials Φ : X → ℝ such that ||Φ|| < ∞ is a Banach Space.
(ii) ℬ₀ ≡ {Φ ∈ ℬ| Φ(X) ≠ 0 only for a finite number of sets 0 ∈ X}, is dense in ℬ

Definition 6.1. A potential $\Phi \in \mathscr{B}^k \subset \mathscr{B}$ satisfying $\Phi(X) = 0$ unless N(X) = k, $k \ge 1$ is known as a **k body potential**.

A potential $\Phi \in \mathscr{B}_0$ is said to have finite range. In the infinite volume limit, translational invariance is an important property of the system, defined as: for all $a \in \mathbb{Z}^d$, $U_{\Phi}(X + a) = U_{\Phi}(X)$ implies $\Phi(X + a) = \Phi(X)$ where $\Phi(\emptyset) \equiv 0$. Translational invariance implies \mathscr{B}^1 is one dimensional, in fact it is easy to see that \mathscr{B}^1 is isometrically isomorphic to \mathbb{R} .

Definition 6.2. A potential $\Phi \in \mathcal{B}$ is a pair interaction if N(X) > 2 implies $\Phi(X) = 0$, i.e. $\Phi = (\Phi^1, \Phi^2) \in \mathcal{B}^1 \oplus \mathcal{B}^2$.

Let Φ be a pair interaction with finite range, i.e. $\Phi \in \mathscr{B}_0^1 \oplus \mathscr{B}_0^2$. Therefore, under this decomposition of \mathscr{B} we have $U_{\Phi}(X) = N(X)\Phi^1(X) + U_{\Phi^2}(X)$ [27]. Therefore

(56)
$$U_{\Phi}(X) = N(X)\Phi^{1} + \sum_{\{x,y\} \subset X} \Phi^{2}(\{x,y\})$$
$$= N(X)\Phi^{1} + \frac{1}{2} \sum_{x \in X} \left[\sum_{\substack{y \in \Lambda \\ y \neq x}} - \sum_{y \in \Lambda - X} \right] \Phi^{2}(\{x,y\})$$
$$\equiv N(X)(\Phi^{1} + C_{\Phi^{2}}) - \frac{1}{2} \sum_{x \in X} \sum_{y \in X^{c}} \Phi^{2}(\{x,y\}).$$

Note that translational invariance of the system implies $\Phi^2\{(x, y)\} = \Phi^2(|x - y|))$ so that $\Phi^2\{(x, y)\} = \Phi^2\{(y, x)\}.$

To clarify the connection with spin systems and, in particular, the classical Ising model with Hamiltonian given in equation (54), it is necessary to remark on the two terms in the last line of equation (56). The 1/2 factor accounts for the double counting performed in the sum decomposition. The term $N(X)(\Phi^1 + C_{\Phi^2})$ is, apart from the negligible constant $\frac{1}{2}V(\Phi^1 + C_{\Phi^2})$, the interaction of the spins with a magnetic field [27]. The last term is the state $1 \leftrightarrow 2$ mutual interactions or "spin up–spin down" $\{\uparrow\downarrow\}$ interactions. With this abstraction of the Ising model, the partition function takes a useful form:

(57)
$$Z_{\Lambda}(\Phi) = \sum_{X \subset \Lambda} \exp\left(-\beta \Phi(X)\right)$$
$$= \sum_{X \subset \Lambda} \exp\left(-\beta N(X)(\Phi^{1} + C_{\Phi^{2}})\right) \exp\left(\frac{\beta}{2} \sum_{x \in X} \sum_{y \in X^{c}} \Phi^{2}(\{x, y\})\right)$$
$$\equiv \sum_{X \subset \Lambda} z^{N(X)} \prod_{x \in X} \prod_{y \in X^{c}} e^{\frac{\beta}{2} \Phi(\{x, y\})} \equiv \sum_{X \subset \Lambda} z^{N(X)} \prod_{x \in X} \prod_{y \in X^{c}} A_{x, y}$$
$$\equiv \mathcal{P}^{n}(z)$$

Theorem 6.2. [27] Let $\{A_{ij}\}_{i\neq j}$ be a family of real numbers such that $A_{ij} = A_{ji}$ and $-1 \leq A_{ij} \leq 1$ for i, j = 1, ..., n. We define a polynomial \mathcal{P}_n of n variables by

$$\mathcal{P}_n(z_1,\ldots,z_n) \equiv \sum_S z^S \prod_{i\in S} \prod_{j\in S^c} A_{ij}$$

where the summation runs over all subsets $S \equiv \{i_1, \ldots, i_s\}$ of $\{1, \ldots, n\}$, $z^S \equiv \prod_{m=1}^s z_{i_m}$, and $S^c = \{j_1, \ldots, j_{n-s}\}$ is the complement of S in $\{1, \ldots, n\}$. Then $\mathcal{P}_n(z_1, \ldots, z_n) = 0$ and $|z_m| \ge 1$ for $m = 1, \ldots, n-1$ implies $|z_n| \le 1$.

Corollary 6.1. (*Lee-Yang*) With the notations and assumptions of theorem 6.2 we define a polynomial \mathcal{P}^n of degree n in z by

$$\mathcal{P}^n(z) = \mathcal{P}_n(z, \dots, z) = \sum_S z^{N(S)} \prod_{i \in S} \prod_{j \in S^c} A_{ij}$$

where N(S) is the number of elements in S. Then the zeros of \mathcal{P}^n all lie on the circle $\{z : |z| = 1\}$

Proof. In the classical ferromagnetic Ising model Hamiltonian, the $\{\uparrow\downarrow\}$ interaction term is positive, $J_{ij} \ge 0$, therefore $\Phi^{(2)} \le 0$. Thus, $\exp(\beta\Phi^{(2)}(\{x,y\})/2) = A_{x,y} \in (0,1]$. Translational invariance implies $\Phi(\{x,y\}) = \Phi(\{y,x\})$ thus $A_{x,y} = A_{y,x}$. Applying theorem 6.2 to $\mathcal{P}^n(z) = 0$ thus implies $|z| \le 1$ and $|z| \ge 1$. Therefore |z| = 1.

Remark 6.1. It should be noted that in theorem 6.1 no assumptions about

- the range of the interaction Φ^2 ,
- the dimensionality of the lattice,
- or the size and structure of the lattice,

are made. Furthermore, the coefficients $A_{x,y}$ do not explicitly dependent on Φ^1 , hence the magnetic field H. This implies the distribution of zeros of the partition function is not explicitly dependent on H. Rather, dependent on the nature of the mutual spin interactions.

The following theorem and its corollary [28] provides an extension of the classical Lee–Yang theorem.

Theorem 6.3. (*Ruelle*) Let \mathcal{P} be a complex polynomial of several variables, which is of degree one with respect to each, i.e. let Λ be a finite set and

$$P(z_{\Lambda}) = \sum_{X \subset \Lambda} c_X z^X,$$

22

where $z_{\Lambda} = (z_x)_{x \in \Lambda}$, $z^X = \prod_{x \in X} z_x$, and $c_X \in \mathbb{C}$. Let Λ_{α} be a finite covering of Λ , and for every $x \in \Lambda_{\alpha}$ let $M_{\alpha,x}$ be a closed subset of \mathbb{C} such that $0 \notin M_{\alpha,x}$. Assume, for each α , that

$$P_{\alpha}(z_{\Lambda_{\alpha}}) = \sum_{X \subset \Lambda_{\alpha}} c_{\alpha,X} z^X \neq 0, \text{ for all } z_x \notin M_{\alpha,x}, x \in \Lambda.$$

Then the polynomial

$$P(z_{\Lambda}) = \sum_{X \subset \Lambda} z^X \prod_{\alpha} c_{\alpha,(\Lambda_{\alpha} \bigcap X)} \neq 0, \text{ for all } z_x \notin -\prod_{\alpha} (-M_{\alpha,x}), x \in \Lambda.$$

Corollary 6.2. (Ruelle)

(i) Lee-Yang Theorem: Let Λ_{α} be the two-point subsets of Λ : $\Lambda_{\alpha} = \{x, y\}$ and $c_{\alpha,X} = A_{xy}$, defined in theorem 6.2, when $X = \{x\}$ or $X = \{y\}$, $c_{\alpha,X} = 1$ when $X = \emptyset$ or $X = \{x, y\}$. For real A_{xy} and $-1 \le A_{xy} \le 1$ we may take $M_{\alpha,x} = \{z \in \mathbb{C} : |z| \ge 1\}$; hence

$$Q(\xi) = \sum_{X \subset \Lambda} \xi^{N(X)} \prod_{x \in X} \prod_{y \in X^c} A_{xy}$$

does not vanish when $|\xi| < 1$. By symmetry $Q(\xi)$ does not vanish when $\xi > 1$, hence the zeros of Q have absolute value 1; this is the Lee-Yang circle theorem.

(ii) **Zero Free Region**: Let Φ be a real function on the d-tuples of integers mod m, the "periodic lattice" $(\mathbb{Z}_m)^d$, with $\Phi(x) = \Phi(-x)$, and take $\Lambda = (\mathbb{Z}_m)^d$. Let Λ_α be the two-point subsets of $\Lambda : \Lambda_\alpha = \{x, y\}$, and write $c_{\alpha,X} = \exp\left[-\beta\Phi(x-y)\right]$ when $X = \{x, y\}$ and $c_{\alpha,X} = 1$ when $X = \emptyset$, $\{x\}$, or $\{y\}$. Take $M_{\alpha,x} = \Delta_{xy}^\beta$ where

$$\begin{split} \Delta^{\beta}_{xy} &= \{z \in \mathbb{C} : |z+1| \leq (1 - e^{\beta \Phi(x-y)})^{1/2}\} \text{ for } \Phi(x-y) \leq 0, \\ \Delta^{\beta}_{xy} &= \{z \in \mathbb{C} : |ze^{-\beta \Phi(x-y)} + 1 \leq (1 - e^{-\beta \Phi(x-y)})^{1/2}\} \text{ for } \Phi(x-y) \geq 0, \\ \text{ Then } \end{split}$$

$$Q(\xi) = \sum_{X \subset \Lambda} \xi^{N(X)} \exp\left[-\beta \sum_{\{x,y\} \subset X} \Phi(x-y)\right]$$

can vanish only when

$$\xi e \Gamma^{\beta} = -\prod_{y \in \mathbb{Z}_m^d} (-\Delta_{0y}^{\beta}).$$

For small β , Γ^{β} does not intersect the positive real axis.

The Lee-Yang theorem implies the partition function and Helmholtz potential can be written in the form

(58)
$$Z = \prod_{i=1}^{N} (z - z_i), \quad z_i = e^{i\theta_i}, \quad A = -\beta^{-1} \sum_{i=1}^{N} \ln|z - z_i|$$

Theorem 6.3 is an extension of the Lee–Yang theorem and can be used to define a region free of zeros on the real axis for temperatures greater than some critical value T_c . This theorem can also be used to show [28] that the zeros \mathcal{Z} remain close to the unit circle even when a small perturbation (possibly many body) is added to the original pairwise interaction. In the infinite volume limit the set of zeros of the partition function are more general than mere point sets [13]. As $N \to \infty$

the average density converges to an analytic function in z both inside and outside the unit circle [36] [27]. Furthermore, $\mathcal{Z} \subset \partial D(0,1)$ in this limit [37] [1]. The distribution of zeros is given by an analytic density function $g(\theta)$ on the unit circle [9]. Theorem 6.3 implies there exists a critical angle $\theta_0(T_c)$ s.t. $z = e^{i\theta}, \theta \in [-\theta_0, \theta_0]$ implies $Z(z) \neq 0$ for $T > T_c$. Therefore [9],

$$\beta A = -\int_{\theta_0}^{2\pi-\theta_0} \ln(z-e^{\mathrm{i}\theta}) dg(\theta).$$

So that the system equilibrium is given by the minimization of a logarithmic potential over measures $g(\theta)$.

7. A Two Parameter Dipole Model of the ER Fluid

When a dielectric ER fluid is in the presence of an external field, the contrast of permittivities between the background fluid and solid particles causes each particle to become polarized with dipole moment aligning with the field [16]. When the particles are far apart, the dipole–dipole interactions dominate. The lowest energy state of two interacting dipoles is one where the separation vector \mathbf{r} and the dipole moments align [16] [15]. This causes the spheres to align, forming chain like structures in the direction of the applied field. Although, as the particles aggregate and touch, higher order multipole–multipole interactions begin to dominate, making local field effects significant. When the applied field is oscillatory, conductivity and/or relaxational effects, influenced by the imaginary part of the contrast parameter, may also be significant [33]. These complications make modeling the ER fluid increasingly difficult.

One of the advantages of using the Bergman–Milton representation (10), is that all of the complicated multi-body interactions are encapsulated in the spectral measure. Although this simplifies things considerably from a theoretical point of view, it does not simplify the analysis of the system, as the spectral measure is in general, incredibly hard to calculate.

The Ising model oversimplifies interactions present in magnetic systems. Yet it captures enough of the behavior that it has become a powerful tool in understanding phase transitions thereof. In this section we present numerical results of a three parameter extension of a two dimensional, square lattice, dipole model, with pairwise interactions. Monte Carlo simulated annealing of the model, illustrates how the body centered tetragonal ground state in three dimensions, might be predicted by a generalization of this two dimensional model.

7.1. The Model and Results. Consider a square lattice assembly of solid dielectric spheres of radius r_s and permittivity ϵ_s , immersed in a background fluid of permittivity $\epsilon_f < \epsilon_s$, subject to an applied field \mathbf{E}_0 in the z-direction. To simplify things, each sphere is modeled by a dipole with dipole moment equal to that of a single sphere, in an infinite background, subject to the uniform field ([16] page 159): $\mathbf{p} = [4\pi r_s^3 \epsilon_f/3(s-2/3)]\mathbf{E}_0$. The interaction energy associated with two dipoles, at a distance r, is $(\mathbf{p}_1 \cdot \mathbf{p}_2 - 3(\mathbf{p}_1 \cdot \hat{\mathbf{r}})(\mathbf{p}_2 \cdot \hat{\mathbf{r}}))/(4\pi \epsilon_f r^3)$ [16] [15]. Therefore the interaction of the modeled dipoles is

(59)
$$\frac{1}{2}\epsilon_f E_0^2 \phi_{ij} = \phi_0 G_{ij}, \quad G_{ij} = \frac{1 - 3\cos^2 \theta_{ij}}{r_{ij}^3}, \quad \phi_0 = \frac{8\pi r_s^6/9}{(s - 2/3)^2}$$

where θ_{ij} is the angle between the dipole moment and the separation vector \mathbf{r}_{ij} . The interaction energy of a single dipole, aligned in the external field, is [15]

(60)
$$U = -\mathbf{E}_0 \cdot \mathbf{p} = -\frac{1}{2}\epsilon_f E_0^2 U_0, \quad U_0 = \frac{8\pi r_s^3/3}{s - 2/3}$$

For reasons mentioned above, this is not an accurate model of the dielectric ER fluid. Although, the geometric factor G_{ij} is negative for $-54.7^{\circ} \lesssim \theta_{ij} \lesssim 54.7^{\circ}$ which creates a bound state, thus giving the correct qualitative behavior. Our model is a three parameter generalization of the above structure, which divides the spatial interactions into three categories, vertical ($\theta_{ij} = 0^{\circ}$), horizontal ($\theta_{ij} = 90^{\circ}$), and other interactions. The system Hamiltonian can be expressed symbolically by

(61)
$$\frac{1}{2}\epsilon_f E_0^2 \mathbf{H} = \sum_{isolated} U_0 + a \sum_{vertical} \phi_{i,j} + b \sum_{horizontal} \phi_{i,j} + c \sum_{other} \phi_{i,j}$$

where a, b, and c are positive parameters used to balance interactions. The summations run over dipole pairs within an interaction range R defined by the condition $\|\mathbf{x}_i - \mathbf{x}_j\|_{\infty} \leq R$. For large R the contribution of of isolated interactions (60) is minimal. The parameter ratios a = c/43, b = c/1.9 and R = 4 generated the sequence of states that lead to the multi-columnar structures shown in figures 2. In these figures, the white squares are the locations of the occupied sites while the black represents the background fluid.

The Monte carlo simulated annealing of this system was performed as follows. The Hamiltonian H_0 is calculated for the initial configuration which has a Gibbs weight of $\exp(-\alpha H_0)$ where $\alpha = \beta \epsilon_f E_0^2/2$. The initial value of the phase transition parameter α is chosen to be $0.01/H_0$, well below a phase transition value. A random occupied site is chosen and the energy of configurations associated with this site shifted to unoccupied nearest neighbor sites, i.e. $\|\mathbf{x}_i - \mathbf{x}_j\|_{\infty} = 1$, is calculated. The probability that the system evolves to one of these shifted configurations is given by the associated Gibbs factor. After the system evolves, allowing the possibility of staying in the current configuration, another random site is chosen for the shifting procedure. This continues until half of the occupied sites have a possibility of being chosen. The value of α is subsequently increased to $\alpha(1 + 2 \times 10^{-10})$ and the procedure repeats. As α increases configurations of lower energy become more probable. The system thus tends to the most probable ground state determined by the Gibbs factor.

It is worth mentioning that this four parameter model can be reduced to an equivalent two parameter model by the following argument. The term in the Hamiltonian which describes the energy associated with isolated dipoles can be omitted. This is because the geometric factor (59) creates a bound state. Therefore, setting U = 0 amounts to isolated dipoles undergoing an unbiased random walk on a random environment until coming within interaction range of other dipoles, eventually forming a bound state for $\alpha \gg 1$. Furthermore, numerical fitting of the model shows that only the ratios of a and b to c are significant. Therefore the parameter c can be absorbed into α leaving the Hamiltonian

(62)
$$\frac{1}{2}\epsilon_f E_0^2 \mathbf{H} = \tilde{a} \sum_{vertical} \phi_{i,j} + \tilde{b} \sum_{horizontal} \phi_{i,j} + \sum_{other} \phi_{i,j}$$

where parameter values $\tilde{a} = \tilde{b} = 1$ produce laminate chain-like structures parallel to the field and parameter values $\tilde{a} = 1/43$, $\tilde{b} = 1.9$ produce the structures shown in figures 2.



FIGURE 2. Various states of the dipole ER fluid as α increases (System size is 35×35 , Interaction range is 4).

Let us briefly recall the classical Ising model, discussed in detail in section 6. The reference measure on the configuration space, $\Omega = \{-1, 1\}^{|\Lambda|}$, associated with the system $\Lambda \subset \mathbb{Z}^d$ is the measure corresponding to the Bernoulli random variable, $\tau = (1/2)^{|\Lambda|}$, as spin-up and spin-down states are equally likely, where d is the dimension, and $|\cdot|$ denotes the cardinality of a set. Therefore, the probability measure for the Ising model is, (see section 6.1):

(63)
$$P(d\omega) = Z_{\Lambda}^{-1}(\beta, H) \exp\left(-\beta \sum_{A \subset \Lambda} \Phi_A(\sigma)\right) d\tau,$$
$$\Phi_A(\sigma) = \begin{cases} J_{i,j}\sigma_i\sigma_j, & \|i-j\| = 1 \text{ for } A = \{i,j\}, \\ H\sigma_i, & \text{ for } A = \{i\}, \\ 0, & \text{ otherwise} \end{cases}$$

where H is the magnetic filed strength, the pairwise interaction strength $J_{i,j} > 0$ $(J_{i,j} < 0)$ for a Ferromagnet (Anti-ferromagnet), σ_i is the standard spin variable (+1 for site *i* being spin-up and -1 for spin-down), and $Z_{\Lambda}(\beta, h)$ is the partition function [27]. It is important to note that many of the results related to the Ising model, including the Lee Yang theorem, do not require the nearest neighbor assumption, rather only require finite range interactions (see section 6.1) [27].

For the two parameter dipole ER fluid model, define lattice gas spin variables: $\mu_i = 1$ for occupied sites, $\mu_i = 0$ for unoccupied sites, and $\sum_{i \in \Lambda} \mu_i \cong |\Lambda| p$ where p is the volume fraction of occupied sites (equality in the infinite volume limit by the law of large numbers). The reference measure for this system is given by

(64)
$$p^{\sum_{i \in \Lambda} \mu_i} (1-p)^{\sum_{i \in \Lambda} (1-\mu_i)} = \exp\left(-h_p \sum_i \mu_i\right) (2(1-p))^{|\Lambda|} (1/2)^{|\Lambda|}$$

where $h_p = \ln((1-p)/p)$. The term $(2(1-p))^{|\Lambda|}$ may be absorbed into the partition function as it is independent of configuration. Therefore, the probability distribution for the dioplar ER fluid model is

(65)
$$P(d\omega) = Z_{\Lambda}^{-1}(\beta, h_p) \exp\left(-\beta \sum_{A \subset \Lambda} \Phi_A(\mu)\right) d\tau,$$
$$\Phi_A(\mu) = \begin{cases} \frac{1}{2} \epsilon_f E_0^2 \tilde{\phi}_{i,j} \mu_i \mu_j & \|i - j\| \le R, \text{ for } A = \{i, j\}, \\ h_p \mu_i, & \text{ for } A = \{i\}, \\ 0, & \text{ otherwise} \end{cases}$$

where $\tilde{\phi}_{i,j} = \tilde{a}\phi_{i,j}$, $\tilde{b}\phi_{i,j}$ for $\theta_{i,j} = 0$, $\pi/2$ respectively (see equation (62)) and $\phi_{i,j}$ is given in equation (59). This model incorporates both ferromagnetic and antiferromagnetic interactions depending on $\theta_{i,j}$. Furthermore, this model has characteristics of a continuous spin Ising model [9].

The potentials Φ_A for both the Ising model and dipole ER fluid model are absolutely summable, translationally invariant, and continuous. These are sufficient conditions to ensure [23]: the probability measures are Gibbs measures, the models have infinite volume limits, an explicit formula for the corresponding infinite volume Helmholtz potential, that the Gibbs measures satisfy a large deviation principle. It is hoped that this abstract framework may give detailed information on ER fluid phase transitions in a similar way as it has for the Ising model (see section 6.1) [23] [27]. If a factorization of the associated partition function can be found, through perhaps a Lee-Yang type theorem, it may also shed light on the nature of these phenomena.

Future numerical work will use the Bergman-Milton representation of the Hamiltonian (see section 5.2) to study the associated simulated annealing. Work done by Dobson, has shown that simulations minimizing the logarithmic potential, corresponding to the Bergman-Milton spectral measure, (the Golden potential), gives chain like structures for dielectrics, and fractal net structures for metals (s = 0). In these simulations, the strength of the electric field was neglected. Therefore, this approach is promising.

8. RANDOM MATRIX THEORY OF DISORDERED SYSTEMS

Random matrices have been used to accurately describe a number of physical phenomena including quantum chaos and disordered mesoscopic conductors (MC) [20]. The mentioned systems have common universal features. Most well known is the result that the probability distribution of level spacings between nearest neighbor eigenvalues in the bulk of the spectrum, demonstrates a transition from a highly correlated Wigner distribution to a Poisson distribution as a function of a parameter that describes the disorder of the system [17]. For disordered MC, the universality of the spacing distribution manifests itself as the universal conductance fluctuation known to exist in the metallic regime. Although, for sufficiently strong disorder, there is a transition from metal to insulator, and the universal conductance

fluctuation, in the insulating regime, is expected to be very different [17]. Why the spectral statistics of these very different systems should be the same is not clear, but there seems to be some kind of Law of Large Numbers hiding in the background.

Numerical simulations of level spacing statistics of the random matrix $\chi_1 \Gamma \chi_1$, at the heart of two component composites (see section 4), exhibits a similar behavior to that of MC, as a function of inclusion volume fraction which determines the disorder of the binary composite. In order to illuminate techniques that might be applied to binary composites, section 8.1 briefly reviews the random matrix theory (RMT) associated with disordered MC. Section 8.2 discusses the results for two component composites, and future research directions.

8.1. Random Matrix Theory of Mesoscopic Conductors. Since the discoverv of the universal conductance fluctuation in MC it has become clear that the conductance is not a self averaging quantity. Therefore, the transport properties of this disordered system should be expressed in terms of the probability distribution of conductance rather than its ensemble average. In particular, due to the fluctuations in the size and shape of the metallic particles, the disorder-induced Anderson transition from a metal to insulator exhibits fluctuations that cannot be explained in terms of the ensemble averaged conductance [17] [34] [35]. One approach which provides a possible framework, for such a system, is based on random transfer matrices T. This matrix gives the flux amplitudes to the right of the conductor in terms of the incoming and outgoing fluxes on the left. In terms of the N non-degenerate eigenvalues $x_n \ge 0$ of the matrix $X = (T^*T + (T^*T)^{-1} - 2)/4$, the conductance is given by $g = \sum_{n} 1/(1+x_n)$ [34] [35]. Using theories of random matrices, the distribution of eigenvalues and its corresponding spacing distribution can be calculated, given the symmetries of X [20]. Such models provide a simple explanation of the universal conductance fluctuations observed in MC in terms of the well-known spectral rigidity characteristic of random matrix ensembles (RME). This approach is especially appealing as it incorporates various symmetries, such as time reversal symmetry breaking in the presence of a magnetic field, in a simple way [17] [34] [35] [20]. The excellent numerical agreement of this approach with the tight-binding Anderson model, and experiments, implicate the general validity of the random matrix model ([35] and references within).

The RME can be derived using a maximum entropy Ansatz of matrices [8] [3]. This method is based on the assumption that a sufficiently complicated system is better described by a matrix which is as random as possible, subject to certain symmetry requirements. To illustrate the general framework, consider a system that may be described by an $N \times N$ matrix X with eigenvalues $a \leq x_i \leq b$, $i = 0, \ldots, N - 1$. The RME of all random matrices X, subject to some physical constraint (e.g., given the average density of eigenvalues) and consistent with given symmetries (Hermiticity, time reversal, etc.), has a distribution of eigenvalues that can be written quite generally in the form [17] [20]

(66)
$$P\{x_0, \dots, x_{N-1}\} \equiv P\{x_i\} = Z^{-1} \prod_{m>n=0}^{N-1} |x_m - x_n|^{\beta} \prod_{k=0}^{N-1} \exp\left(-V(x_k)\right).$$

Here β is a parameter depending only on the symmetries of X and is equal to 1, 2, or 4 for orthogonal, unitary, and symplectic symmetries, and Z is the normalization constant. One can think of equation (66) as an exponential of some effective

"Hamiltonian" H for the eigenvalues by writing $P = Z^{-1} \exp(-\beta H)$, where

(67)
$$H\{x_0, \dots, x_{N-1}\} = \beta^{-1} \sum_{k=0}^{N-1} V(x_k) - \sum_{m>n=0}^{N-1} \ln |x_m - x_n|.$$

The first term corresponds to a single-particle confining potential which keeps the eigenvalues from moving out to infinity. This competes against the second term which corresponds to a logarithmic repulsion between the eigenvalues which try to keep them far apart from each other. Different models within the same symmetry class correspond to different forms of the single particle potential V while keeping the interaction term the same. In the language of the maximum entropy Ansatz, the single particle potential is a Lagrange multiplier function that fixes, e.g., the eigenvalue density (corresponding, e.g., to a given mean value of conductance) [17].

The distribution of conductance g can be obtained from $P\{x_i\}$ as follows [34] [35]:

(68)
$$P(g) = \left\langle \delta\left(g - \sum_{n=0}^{N-1} \frac{1}{1+x_n}\right) \right\rangle,$$

where $\delta(x)$ denotes the Dirac delta distribution. Respectively, the average and partition function is given by

$$\langle (\cdots) \rangle = Z^{-1} \int_{[a,b]^N} \left(\prod_{n=0}^{N-1} dx_n \right) P\{x_i\}(\cdots), \quad Z = \int_{[a,b]^N} \left(\prod_{n=0}^{N-1} dx_n \right) P\{x_i\}.$$

For an explicit form of V, we define a family of polynomials $P_n(x)$ orthogonal to the weight $\exp(-V)$:

(70)
$$\int_{a}^{b} dx \ e^{-V(\lambda)} P_n(x) P_m(x) = \delta_{n,m} h_n.$$

If the polynomials are known explicitly, it is possible to calculate any *n*-point correlation function for this distribution *exactly*, for any given N [20]. The corresponding two-point kernel $K_N(x, y)$ is defined as

(71)
$$K_N(x,y) = e^{-(V(x)+V(y))/2} \sum_{n=0}^{N-1} P_n(x) P_n(y) / h_n.$$

The eigenvalue density and spacing distribution can then be calculated in terms of this kernel. Indeed, for translationally invariant kernels, the spacing t between nearest eigenvalues, in units of average local spacing, for large N, has the spacing distribution p(t) given by [20]

(72)
$$p(t) = \frac{d^2 E(t)}{dt^2}, \quad E(t) = \det(1 - \bar{K}_t)$$

where \bar{K}_t is the kernel divided by the local density and restricted to an interval of length t [17].

The transition of the level spacing distribution, from Poisson to Wigner, has been accurately described numerically by a single particle potential V that has the following asymptotic behavior: $V(x) \sim (\ln x)^2$ for $x \gg 1$ and $V(x) \sim x$ for $x \ll 1$ [34] [35]. Consider the potential given by

(73)
$$w(s;q) = \exp(-V(x;q)) = x^{\alpha}/(-(1-q)tx;q)_{\infty}$$
$$(a;q) = \prod_{n=0}^{\infty} (1-aq^n), \quad 0 < q < 1, \ \alpha > -1.$$

It can be shown that V(x;q) has a power series expansion in x for $x \ll 1$, $\alpha = 0$ and as $q \to 1^-$, $w \to e^{-x}$ giving the Wigner level spacing distribution. On the other hand, for $x \gg 1$, $q \ll 1$ the single particle potential $V(x) \sim (\ln x)^2$ giving the Poisson level spacing distribution. Therefore, the single particle potential (73) has the required asymptotic behavior to accurately describe the metal/insulator transition observed in MC. It is to be expected that in general, t, α , and q will depend on physical parameters such as disorder, size, and dimension. The model is exactly solvable because, for the potential given in equation (73), the polynomials are known explicitly for the entire parameter range [34]. They are the q-Laguerre polynomials $L_N^{(\alpha)}(x;q)$. In terms of these polynomials, the normalized eigenvalue density is [34]

(74)
$$\frac{\sigma_N(x;q)}{N} = w(x;q) \sum_{k=0}^{N-1} \frac{[L_k^{(0)}(x;q)]^2}{h_k}, \quad h_k = \ln(1/q)/(1-q)q^k.$$

In the limit $q \to 1^-$, $L^{(0)}(x;q)$ reduces to the ordinary Laguerre polynomials [17]. In this limit, the confining potential dominates the logarithmic interaction term, at large x, thereby forcing the eigenvalues towards the origin. This gives the large number of near zero spacings seen in the Poisson distribution. Using properties of Laguerre polynomials, it is known that the variance of g is independent of N and t, giving rise to the universal conductance fluctuation. Asymptotic analysis of the case q = 1 describes the metallic regime quite well [34] [35]. For $t \to \infty$ all eigenvalues are compressed at the origin giving $\langle g \rangle = N \gg 1$. In the large N limit and for $Nt \gg 1$ the asymptotics reproduce Ohm's law and gives the leading "quantum" correction in 3D. On the other hand, for $q \ll 1$, the large-x single particle potential, being essentially logarithmic, is very weakly confining, allowing the eigenvalues to spread out to the point that the density becomes discrete near the origin even in the thermodynamic limit [35]. In this regime, the q-Laguerre polynomials have an asymptotic expansion qualitatively different from that of q = 1. Furthermore, for q < 1 the measure w is not unique. In this case there exist many different but equivalent measures with the same polynomials and moments. The classic moment problem shows that the extreme measure, associated with this equivalence class of measures, is discrete and is supported at the zeros of an entire function. The explicit from for a representative measure is $w_{dis}(x;q) = \sum_{n=0}^{\infty} w_n \delta(x-\tau_n)$, where τ_n are the zeros of $L_{\infty}^{(\alpha)}(x;q)$ and the $\{w_n\}$ can be explicitly determined in terms of x derivatives of $L_{\infty}^{(\alpha)}(x;q)L_{\infty}^{(\alpha)}(x/q;q)$ evaluated at $x = \tau_n$ [35]. Asymptotic analysis shows that the eigenvalue density for $\alpha = 0$ is given by

(75)
$$\sigma_{\infty}(x;q) = \frac{1-q}{\ln(1/q)} \sum_{n=0}^{\infty} \delta(x-\tau_n) \approx \begin{cases} 0, & x(1-q) \le 1/q\\ (1-q)/(2\gamma) & x(1-q) > 1/q \end{cases}.$$

where $\gamma = (-\ln q)^x$ is the measure of disorder [35]. This shows that the appearance of a gap is a clear indicator of an insulator. Indeed, from the definition of $\langle g \rangle$ one

can show, for $q \ll 1$, the mean conductance is exponentially small. Specifically, as the localization length becomes smaller than the system size, q changes from 1 to a value smaller than 1 and the eigenvalue density at the origin gets depressed. A gap opens up, at sufficiently small q, indicating a metal to insulator transition. It is important to note, that although the discrete measure w_{dis} is quite distinct form the continuum measure w, their asymptotics are essentially the same and both give identical results [34].

8.2. Random Matrix Theory of Two Component Conductors. In order to parallel the discussion of MC and appeal to intuition associated with conductors we discuss our results in terms of the metal/insulator transition of percolating two component random resistor networks (RRNs). The mathematical equivalence of this system to random capacitor networks (RCN) [11] (see section 4) requires that the discussion of RRN has a natural analogue for RCN.

To briefly put the system discussed in section 4 in the language of RRNs, consider a two component bond lattice RRN with conductance $\sigma = \sigma_1 \chi_1 + \sigma_2 \chi_2$ where $\sigma_1 < \sigma_2$, χ_1 is the characteristic function of component one, $\chi_2 = 1 - \chi_1$. A given resistor has probability $p = \langle \chi_2 \rangle_V$ of being a type two resistor and probability 1-pof being the other type, where $\langle \cdot \rangle_V$ denote the volume average. Thus, the geometric aspects of the system, in particular the disorder, is completely determined by the volume fraction p. When the contrast ratio $h = \sigma_1/\sigma_2$ is zero (s = 1) we can view the conductivity zero bonds as being vacant. In this case, the underlying percolation problem is described in terms of $P_{\infty}(p)$, the probability that the origin is connected to an infinite cluster of occupied sites. For p below some critical probability p_c , called the percolation threshold, $P_{\infty}(p) = 0$ while for $p > p_c$, $P_{\infty}(p) > 0$. The effective conductivity has a similar behavior: $\sigma^*(p) = 0$ for $p < p_c$ and $\sigma^*(p) > 0$ for $p > p_c$ [12].

The percolation aspects of this system are determined by the spectral measure of the real symmetric random matrix $\chi_1\Gamma\chi_1$ which depends only on the volume fraction p, the dimension, the topology of the underlying lattice, and boundary conditions for finite systems. The same analysis done in section 4 can be applied to two component RRNs [12] giving the effective conductance σ^* in terms of the eigenvalues of this matrix. The result is the Bergman-Milton representation of the effective conductance.

(76)
$$\frac{\sigma^*(p)}{\sigma_2} = 1 - \sum_i \frac{\mu_i}{s - \lambda_i}$$

Hence the discrete spectral measure is given by $d\mu(\lambda) = \sum_i \mu_i \delta(\lambda - \lambda_i) d\lambda$ where $\{\lambda_i\} \equiv \{\lambda_0, \ldots, \lambda_{n-1}\}$ are the eigenvalues of the random matrix and $\{\mu_i\}$ are the masses of the measure.

8.2.1. Statistical Mechanics Approach. When viewing the figures discussed in section 8.2.2, one can see that the RME associated with the $n \times n$ real symmetric random matrix $\chi_1 \Gamma \chi_1$ closely parallels that of MC for $p < p_c$. Therefore, assuming that there exists a mapping from one system to the other, we may write the eigenvalue distribution for this random matrix as $P_n\{\lambda_i\} = Z_n^{-1} \exp(-\beta H_n) d^n \lambda$ with Hamiltonian $H_n\{\lambda_i\} = \beta^{-1} \sum_{k=1}^n V(\lambda_k) - n^{-1} \sum_{m>k=0}^n \ln |\lambda_m - \lambda_k|$ and partition function $Z_n = -n \sum_{k=1}^n V(\lambda_k)$, where β plays the role of a coupling constant for the n-particle system which is determined by symmetries associated with the system and $d^n \lambda = d\lambda_1 \cdots d\lambda_n$ is the Lebesgue product measure on [0, 1] [22]. By

assumption the potential V is related to that given in (73) through a change in variables. The parameter q, related to disorder, must be a function of volume fraction q = q(p), and the scaling n^{-1} plays the role of temperature and leads to a non-random infinite volume limit. This section is devoted to the exploration of the statistical mechanics structure of this RME.

A quantity that is of considerable interest in RMT is the normalized counting measure (NCM) [22] corresponding to the eigenvalues $\lambda_1^{(n)} \leq \lambda_2^{(n)} \leq \ldots \leq \lambda_n^{(n)}$ of $\chi_1 \Gamma \chi_1$: $N_n(\Delta) = n^{-1} | \{\lambda_i : \lambda_i \in \Delta \subset [0,1] \} | = \int_{[0,1]^n} \sum_i n^{-1} \delta_{\lambda_i} d^n \lambda$, where $| \cdot |$ denotes the cardinality of a set and δ_{λ} is the Dirac measure centered at λ . Two related quantities of interest are its expectation value, $E\{N_n(\Delta)\}$, and its variance, $D_n = E\{N_n^2(\Delta)\} - E^2\{N_n(\Delta)\}$ [22]. The NCM is related to the Bergman-Milton spectral measure since $\mu(\lambda) = \sum_i \mu_i \delta_{\lambda_i}$ and therefore have identical supports.

Theorem 8.1. Let the potential V satisfy the following conditions

$$V(\lambda) \ge (2+\delta) \ln |\lambda|$$

for $|\lambda| \ge L$, $\delta > 0$, and $L < \infty$. Furthermore let

(77)
$$|V(\lambda_1) - V(\lambda_2)| \le K |\lambda_1 - \lambda_2|^{\gamma}$$

for $|\lambda_1|, |\lambda_2| \leq L, \gamma > 0, K < \infty, L \leq \infty$. Then the NCM of this RME converges in probability to the non-random, i.e. $D_n \to 0$, absolutely continuous integrated density of states (IDS): $N(\Delta) = \lim_{n\to\infty} E\{N_n(\Delta)\} = \int_{\Delta} \rho(\lambda) d\lambda$. The density of states (DOS) $\rho(\lambda)$ is uniquely determined by the conditions $\rho(\lambda) \geq 0, \int_0^1 \rho(\lambda) d\lambda = 1$, such that $-\int_0^1 \int_0^1 \ln |\lambda - \lambda'| \rho(\lambda) \rho(\lambda') d\lambda d\lambda' \equiv -\int_0^1 U(\lambda) \rho(\lambda) d\lambda < \infty$, where the function $u(\lambda) = U(\lambda) - V(\lambda)$ must be bounded from above, and we must have the support property $supp(\rho) \subset \{\lambda : u(\lambda) = \sup_{\lambda'} u(\lambda')\}$ [22].

This result can be used to show that the ground state energy of the statistical mechanics model, i.e. $\mathcal{E} = \lim_{n \to \infty} n^{-2} \ln Z_n$, has the following form [22]:

(78)
$$\mathcal{E} = \frac{\beta}{2} \int_0^1 \int_0^1 \ln|\lambda - \lambda'|\rho(\lambda)\rho(\lambda')d\lambda d\lambda' + \int_0^1 V(\lambda)\rho(\lambda)d\lambda$$

where $\rho(\lambda)$ is the DOS given in theorem 8.1. Moreover, in total agreement with statistical mechanics, the ground state is obtained by the minimum value of the "electrostatic" energy of two dimensional (line) charges whose distribution on [0, 1] is described by the probability measure ν given by

(79)
$$\mathcal{E}[\nu] = \inf_{\eta} \left\{ -\frac{\beta}{2} \int_0^1 \int_0^1 \ln |\lambda - \lambda'| d\eta(\lambda) d\eta(\lambda') + \int_0^1 V(\lambda) d\eta(\lambda) \right\}$$

Theorem 8.1 then implies [22] that, under its conditions, a minimizing measure has a density satisfying the conditions of this theorem [22]. Therefore, by uniqueness, this density is equal to the DOS $\rho(\lambda)$. The minimizing measure in equation (79) is also the unique solution of the extremum equation of the variational problem

(80)
$$\beta \int_{supp(\rho)} \ln |\lambda - \lambda'| \nu(\lambda') d\lambda' = V(\lambda) + c, \quad \lambda \in supp(\rho) = supp(\mu).$$

where c is a constant [22]. Differentiation of equation (80) with respect to λ yields the singular integral equation

(81)
$$V'(\lambda) = \beta \int_{supp(\rho)} \frac{\rho(\lambda')d\lambda'}{\lambda - \lambda'}, \quad \lambda \in supp(\nu)$$

valid for any function $V(\lambda)$ satisfying the conditions of theorem 8.1.

The conditions on V for which this framework holds has been greatly generalized [30]. Equation (81) has the electrostatic interpretation of the equilibrium condition for continuously distributed charges on [0, 1] of strength $\beta^{1/2}$ subjected the electrostatic potential. The fact that $supp(\rho) = supp(\mu)$ suggests that the Bergman-Milton spectral measure might also satisfies equations 78- 81 which would be very useful for characterizing the ground states of the ER fluid.



FIGURE 3. Eigenvalue Statistics $(p = 5 \times 10^{-5}, N = 350)$

8.2.2. Disorder Dependent Phase Transitions in Binary Composites. The following preliminary figures were obtained from an extension of a program written by Dobson. This program calculates the spectral measure of $\chi_1 \Gamma \chi_1$ for a two dimensional square $N \times N$ bond lattice with periodic boundary conditions in both directions, i.e. a checkerboard on a torus. This program was used as a black box to calculate the eigenvalue statistics of the random matrix of interest. The graphs are linearly connected points representing the normalized number density of events for each bin.



FIGURE 4. Eigenvalue Statistics (p=0.1, N=20)

It can be seen in figure 3 that for low volume fraction, $p \ll 1$, the eigenvalue density $P(t = \lambda)$ is symmetric and highly localized about its mean t = 1/2 creating large spectral gaps about the spectral end points. It is so localized that we conjecture that as $p \to 0$ the small side lobes of the density will diminish causing $P(\lambda = t) \rightarrow \delta(t - 1/2)$ in the limit. The symmetry of the eigenvalue density may be a general feature of the random matrix $\chi_1 \Gamma \chi_1$. This may be understood by recalling that singular behavior of a percolating normal/superconductor composite (s = 0)and insulator/conductor composite (s = 1) is determined by the same random matrix. This matrix is completely determined by the geometry and volume fraction of inhomogeneities therefore is independent of s. This volume fraction regime is analogous to the q = 1 limit in the random matrix theory (RMT) of mesoscopic conductors (MC). There, the confining potential $V(x) \sim x$ dominates the logarithmic repulsion potential, thereby forcing all the eigenvalues to the origin [34] [35]. This greatly increases the number of near zero level spacings which causes the level spacing distribution to exhibit Poisson-like characteristics. A Poisson level spacing distribution corresponds to uncorrelated spectra [17] which suggests that the system itself is uncorrelated in this regime. This is the same behavior demonstrated by the spectra of $\chi_1 \Gamma \chi_1$, subject to the symmetry mentioned, suggesting that the same underlying mechanism might be at work (see figure 3).



FIGURE 5. Eigenvalue Statistics (p=0.3, N=20)

As the volume fraction increases the variance of the spectral density also does, depressing the density at t = 1/2 and decreasing the spectral gaps. The symmetry persists as the side lobes become very pronounced including small lobes near the spectral endpoints starting a build up of spectrum there (see figure 4). This transition manifests itself in the eigenvalue spacing density as an increase in larger spacings creating an increasingly long tail giving transitioning Poisson-Wigner characteristics. This trend continues until the density becomes more and more uniform in the bulk of the spectrum, while the lobes near the spectral endpoints become very pronounced further narrowing the spectral gaps (see figure 5). The affect on the spacing distribution is an increasingly long tail creating a level spacing distribution which greatly resembles the Wigner distribution [17] (see figure 5). A Wigner spacing distribution corresponds to highly correlated spectra [17] which suggests that the long range connectedness in the binary composite for this parameter range causes long range correlations in the spectrum. This behavior is also demonstrated

34

in the RMT of MC. Specifically [34], as the localization length becomes smaller than the system size, q changes from 1 to a value smaller than 1 and the eigenvalue density at the origin gets depressed. A gap opens up, at sufficiently small q, indicating a metal to insulator transition.



FIGURE 6. Eigenvalue Statistics (p=0.4, N=20)

When the volume fraction is increased to about p = 0.38 the spectral gaps collapse symmetrically leading to singular behavior of the effective conductivity for metal (s = 0) or insulator (s = 1) constituents. As the volume fraction continues to increase, the eigenvalue density becomes more and more uniform in the bulk of the spectrum, piling up at the spectral endpoints. This manifests itself in the eigenvalue spacing distribution as the instability shown in figures 6-7 leading to the sharp increase of near zero spacings seen in figure 8, giving rise to a third system behavior. It is expected that the eigenstatistics have three distinct regimes corresponding to $p < p_c$, $p \sim p_c$ and $p > p_c$. As this trend continues, more and more eigenvalues pile up at the spectral endpoints while the bulk of the spectrum becomes virtually uniform. This causes the level spacing distribution to transition to a Poisson-like distribution with an extremely long tail (see figure 9). This makes qualitative sense as the system formulation, that leads to the random matrix being studied, is symmetric in $(\epsilon_1, \chi_1), (\epsilon_2, \chi_2)$. Therefore, we conjecture that $\lim_{p\to 0^+} P(\lambda_{i+1} - \lambda_i = t)$ $= \lim_{p\to 1^-} P(\lambda_{i+1} - \lambda_i = t)$, in some perhaps unbounded sense (see below).

As the volume fraction tends to unity, the eigenvalue density does not accumulate to Dirac point masses at the spectral endpoints, rather diverging with some power law. This manifests itself in the spacing distribution by a power law divergence of near zero spacings in the infinite volume limit. This may lead to the nonexistence of the partition function in the infinite volume limit, i.e. violation of normalization. Such a quasi-Poisson behavior that lead to a new qualitative feature in the RMT of MC, responsible for a violation of normalization and the existence of finite "level compressibility," is the *multifractality* of the eigenvectors [19]. In MC the multifractality is related to the overlapping of different wave functions with different energies. This suggest that, in the infinite volume limit, for $p \sim 1$ the orthogonal eigenspaces associated with the real symmetric matrix $\chi_1 \Gamma \chi_1$ might overlap in some fractal dimension. A generalization of the RMT framework for MC has been developed [18] that explains all three transport regimes in MC: metallic,



FIGURE 7. Eigenvalue Statistics (p=0.42, N=20)

insulating, and critical. This development makes the prospect of a RMT approach that gives a full description of two component composites very promising.



FIGURE 8. Eigenvalue Statistics (p=0.45, N=20)



FIGURE 9. Eigenvalue Statistics (p=0.5, N=20)

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