Lagrangian treatment of magnetic dielectrics

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A general Lagrangian-based long-wavelength theory of ordered magnetic dielectric crystals (ferromagnetic, antiferromagnetic, and ferrimagnetic) is formulated. Our classical treatment of intrinsic spin uses the anticommuting Grassmann algebra G_3 developed by Berezin and Marinov and by Casalbuoni. The Grassmann formulation of classical spin gives by the Dirac quantization procedure the usual nonrelativisitic spin-$\frac{1}{2}$ quantum theory. The treatment begins at the microscopic level before a long-wavelength limit is performed to obtain a macroscopic theory so as to incorporate all long-wavelength modes of motion (acoustic, optic, electromagnetic, and spin) and their interaction to all orders of nonlinearity. The crystals can have any symmetry, anisotropy, and structural complexity. The equations of motion for all the modes are obtained and the energy, momentum, pseudomomentum, angular momentum, and spin conservation laws are found. The magnetizations arising from spin and from the motion of bound charge are found to enter the energy conservation law distinguishably. This theory should be particularly useful for the study of magneto-optical phenomena.

I. INTRODUCTION

Many magnetic phenomena such as magnetoelastic interactions, spin waves, and magneto-optic interactions are long-wavelength phenomena in which the medium can be considered a continuum provided, of course, that all relevant excitations are represented by their own continuum fields. Derivations of such macroscopic phenomena from quantum mechanical beginnings and from classical beginnings must agree. The choice of approach thus depends on issues such as ease and generality. For phenomena in dielectric crystals involving interactions, linear and nonlinear, of acoustic, optic, and electromagnetic modes, a classical Lagrangian approach[1,2] has attained a very general, fundamental, and heuristic formulation. This is illustrated by its use in reexamining both supposedly well-known interactions (the elasto-optic effect,[3] electrostriction,[4] acoustic harmonic generation in piezoelectric crystals,[5] Cauchy symmetry of the stiffness tensor,[6] mechanisms of optical activity[7]), in offering resolutions to long standing controversies (the momentum density of a light wave in a dielectric,[8] the additional boundary condition problem of exciton polaritons[9]), and in exploring new higher-order interactions (acoustically induced optical harmonic generation,[10] a five-wave triply phase-matched acousto-optic interaction,[11] creation of counterpropagating subharmonic ultrasonic waves by microwave excitation[12]). Because of these varied successes it is worth expanding this Lagrangian-based approach to include the intrinsic spin degree of freedom in order to handle ferromagnetic, antiferromagnetic, and ferrimagnetic phenomena and their interactions with acoustic, optic, and electromagnetic excitations.

The incorporation of a spin source of magnetization into a classical treatment of continuum, long-wavelength magnetic phenomena has always presented problems. Somehow spin is not a classical quantity. This is particularly evident when approached from a Lagrangian point of view. The matter portion of a continuum Lagrangian is a difference of kinetic and potential energies. The derivatives in an Euler-Lagrange equation operate on the kinetic energy terms to produce a second-order differential equation in time (a Newton equation) for each degree of freedom. However, it is well known that the magnetization, or the spin creating it, obeys a *first-order* differential equation in time, one in which the time derivative of spin is proportional to the vector product of the spin and an effective magnetic field. Thus a kinetic energy of spin is not a meaningful quantity, a fact also apparent from a quantum mechanical Hamiltonian of spin. The dynamic replacement for the kinetic energy needed in a classical Lagrangian is far from apparent.

Since the dynamic equation for spin (or the magnetization it produces) has the generic form of the time rate of change of a vector whose magnitude remains fixed, previous continuum treatments[13–18] of magnetic phenomena have derived its form with the use of the phenomenological assumption of a fixed magnitude of macroscopic saturated magnetization. Most of these[13–17] avoided a Lagrangian approach, instead using the virtual work principle,[13,14] postulation of the forms of the conservation laws,[15,16] or the virtual power principle.[17] The Valenti-Lax work[18] is Lagrangian based. It uses a combination of the assumed fixed magnitude of magnetization, a nonrotationally invariant "gyroscopic term" in the Lagrangian that involves a set of "external" vectors, and the magnetization as a Lagrangian variable. The external vectors drop out of the torque equation for spin and rotational invariance is restored. The theory also can be quantized. In spite of those successes its unusual basis raises the question whether some other Lagrangian formulation can be found.

There are also a few phenomenological Lagrangian theories that start from assuming two kinetic terms for spin and use spin as a Lagrangian variable without introducing the "external" vectors.[19,20] These theories have recently drawn interest in application to frustrated antiferromagnetic media.[21,22] These theories yield the well-known magnetic-torque equation only by imposing the constraint of constant magnitude of the magnetization variable. There has been no discussion about their canonical formalisms and their quantizations do not appear to yield the well-known quantum mechanical theory for spin.

We believe that the fundamental questions about a classical Lagrangian formulation of intrinsic spin were answered rather recently by the work of Berezin and Marinov[23] and of Casalbuoni.[24] They showed using the coherent state representation of quantum mechanics that the classical limit of a bosonic operator is an ordinary classical quantity but that the classical limit of a quantum mechanical fermionic operator is an anticommuting, but otherwise classical quantity. This explains why Lagrangians have easily included bosonic excitations, such as photons, acoustic phonons, and optic phonons, and why fermionic excitations have been excluded. Clearly, classical physics has been limited by its exclusion of anticommuting variables, the subject of Grassmann algebra.

The cited works[23,24] develop a full canonical formalism for a Grassmann variable and show that the Dirac quantization procedure[25] gives back a quantum theory of fermions. In particular they show that the Grassmann algebra G_3, whose elements form a real three-component anticommuting vector, when quantized, yields the nonrelativistic spin-$\frac{1}{2}$ quantum theory. The canonical formalism produces a classical Lagrangian for a particle possessing spin $\frac{1}{2}$. This is readily incorporated into our Lagrangian formulation for magnetic crystals because we start at the microscopic classical particle point of view before passage to a long-wavelength (continuum) limit. Of course, in ferromagnetic materials the "spin" of an ion is often greater than $\frac{1}{2}$. Since nonrelativistic quantum mechanical theories of spin higher than $\frac{1}{2}$ have the same structure, we surmise that the spin-$\frac{1}{2}$ theory with an altered gyromagnetic ratio can account adequately for a higher spin. The canonical formalisms of Grassmann algebras that presumably correspond to spin-$\frac{3}{2}$, -$\frac{5}{2}$, etc. particles have not been developed as yet.

In this paper we incorporate the Berezin-Marinov-Casalbuoni handling of intrinsic spin into our Lagrangian-based theory of interactions in dielectric crystals. Section II introduces the Grassmann algebra, the canonical formalism of a Grassmann variable, and the Dirac quantization procedure[25] for such a variable. The latter gives credence to the entire formalism and also serves to motivate the definition of spin in terms of Grassmann variables. The remaining formulation of the Lagrangian is presented as briefly as possible since its conceptual and manipulative background has been presented in detail before.[2] The Lagrangian includes all long-wavelength modes of mechanical motion, that is, acoustic modes, optic modes, and now spin modes, in interaction with the electromagnetic field. The crystalline dielectric medium can have any symmetry, anisotropy, and structural complexity. Nonlinearity of any order in the interactions between any of the excitations is included, being limited only by the requirements of satisfying the conservation laws of momentum, angular momentum, and energy. Since we consider a homogeneous medium (before excitation), we also present the pseudomomentum conservation law and, of course, the magnitude of spin is also conserved. The Lagrangian is constructed in Sec. III, the equations of motion are presented in Sec. IV, and the conservation laws are given in Sec. V. Emphasis is placed on the spin additions and modifications throughout.

We believe that the previous applications of this Lagrangian approach to a wide range of phenomena provide ample justification for its further generalization to include magnetic phenomena resulting from intrinsic spin. Further, we believe its mode of introduction of spin is novel and heuristic. We believe this formulation will be particularly useful for the study of magneto-optical phenomena because of its inclusion of all optic modes which provide the dispersion to optical properties. Previous continuum formulations[13–18] do not contain the optic modes of motion while quantum mechanical formulations typically lack the generality of this formulation.

II. GRASSMANN ALGEBRA AND CLASSICAL SPIN VARIABLES

In this section, we distill the relevant contents of Refs. 23 and 24 in order to provide a systematic and complete introduction of the Grassmann algebra and classical spin dynamics relevant to continuum magnetism. It is sufficient for the purpose of this section to consider a single particle. First, we introduce the definition of the Grassmann variables and their algebraic and differentiation rules. Second, we deduce proper definitions for Poisson brackets involving ordinary variables and Grassmann variables that give the correct form to Hamiltonian dynamics and follow a defined property over an algebraic ring. Third, we introduce the action for a nonrelativistic spin-$\frac{1}{2}$ particle and deduce its canonical variables. Fourth, we follow Dirac's procedure to quantize the classical theory. Finally we show that the algebraic generators of the infinitesimal canonical transformation for a three-component Grassmann vector give rise to the nonrelativistic spin $\frac{1}{2}$ and that they become the spin operators represented by Pauli matrices after quantization. We also derive the well-known quantum dynamical equation of spin from its classical counterpart, which shows the dual formalism of the quantum spin theory and the classical mechanics of spins represented through Grassmann variables.

A. Grassmann algebra, canonical formalism, Poisson brackets, and quantization

Grassmann variables are real anticommuting variables defined by

$$[\xi_i, \xi_j]_+ \equiv \xi_i \xi_j + \xi_j \xi_i = 0 \quad (i = j \text{ or } i \neq j), \tag{2.1}$$

where i and j are three-dimensional vector indices and $[,]_+$ is an anticommutator. Note that the internal degrees of freedom represented by a Grassmann vector can be higher than 3, in which case it may represent a relativistic spin-$\frac{1}{2}$ or other higher-spin particles. Since we study only the nonrelativisitic spin-$\frac{1}{2}$ case here and use the three-component Grassmann vector, we choose its component indices to be the three-dimensional vector indices for simplicity. The right derivative with respect to the Grassmann variable is used, that is,

$$\delta f(\xi_j) = \delta \xi_i \partial f(\xi_j)/\partial \xi_i. \tag{2.2}$$

We assume that the dynamics of a classical system can be described by the action principle, with an action given by

$$S = \int L\left(q_i, \dot{q}_i; \xi_i, \dot{\xi}_i\right) dt, \tag{2.3}$$

where q_i are conventional c-number variables, representing mechanical degrees of freedom of the particle. From a general variation of the action we get

$$\delta L = \delta q_i \frac{\partial L}{\partial q_i} + \delta \dot{q}_i p_i + \delta \xi_i \frac{\partial L}{\partial \xi_i} + \delta \dot{\xi}_i \pi_i, \tag{2.4}$$

where the canonical momenta are defined by

$$p_i \equiv \frac{\partial L}{\partial \dot{q}_i}, \quad \pi_i \equiv \frac{\partial L}{\partial \dot{\xi}_i}. \tag{2.5}$$

Generally speaking, functions that contain products of Grassmann variables can be divided into two classes, even functions having an even number of Grassmann factors and odd functions having an odd number of Grassmann factors. They then have the properties

$$[E_1, E_2] = [E, O] = 0, \quad [O_1, O_2]_+ = 0, \tag{2.6}$$

where E_1, E_2, E and O_1, O_2, O denote even and odd functions, respectively, and $[,]$ is a commutator. Since a canonical transformation can mix the characters of generalized coordinates and momenta, they must have the same commuting properties. Thus $\boldsymbol{\pi}$ must be an anticommuting Grassmann variable because $\boldsymbol{\xi}$ is. Therefore the Lagrangian must be even in the Grassmann variables. In the three-component Grassmann vector case, the Lagrangian must be quadratic in Grassmann variables and the canonical momentum must be a Grassmann variable. Thus for such a system we must have the properties

$$[L, \xi_i] = 0, \quad [L, \pi_i] = 0, \tag{2.7}$$

$$[\xi_i, \pi_j]_+ = [\pi_i, \xi_j]_+ = 0, \quad [\pi_i, \pi_j]_+ = 0. \tag{2.8}$$

By setting the variation of the action Eq. (2.3) to zero, using Eq. (2.4), and using the independence of the variations δq_i and $\delta \xi_i$, we obtain the usual Euler-Lagrange equations

$$\frac{d}{dt}\frac{\partial L}{\partial \dot{q}_i} = \frac{\partial L}{\partial q_i}, \quad \frac{d}{dt}\frac{\partial L}{\partial \dot{\xi}_i} = \frac{\partial L}{\partial \xi_i}. \tag{2.9}$$

By the usual procedure and with the use of the right derivative of Eq. (2.2) the Hamiltonian is defined as

$$H = \dot{q}_i p_i + \dot{\xi}_i \pi_i - L. \tag{2.10}$$

We can calculate the variation of H analogous to that of the Lagrangian L to obtain the Hamilton equations

$$\dot{p}_i = -\frac{\partial H}{\partial q_i}, \quad \dot{q}_i = \frac{\partial H}{\partial p_i}, \tag{2.11a}$$

$$\dot{\pi}_i = -\frac{\partial H}{\partial \xi_i}, \quad \dot{\xi}_i = -\frac{\partial H}{\partial \pi_i}. \tag{2.11b}$$

Note that there is a sign difference between the Hamilton equations for $\dot{q}_i$ and $\dot{\xi}_i$. This is the direct result of the anticommuting property of Grassmann variables.

By using Eqs. (2.11) we can evaluate the time derivative of a general function of the canonical variables

$$\frac{d}{dt} Z(q_i, p_i; \xi_i, \pi_i; t) = \frac{\partial Z}{\partial t} + \left(\frac{\partial H}{\partial p_i}\frac{\partial Z}{\partial q_i} - \frac{\partial H}{\partial q_i}\frac{\partial Z}{\partial p_i}\right) - \left(\frac{\partial H}{\partial \pi_i}\frac{\partial Z}{\partial \xi_i} + \frac{\partial H}{\partial \xi_i}\frac{\partial Z}{\partial \pi_i}\right). \tag{2.12}$$

By assuming that the classical Liouville equation

$$\frac{dZ}{dt} = \frac{\partial Z}{\partial t} + \{Z, H\} \tag{2.13}$$

holds here, the Poisson brackets can be extended to include the Grassmann variables. However, we have not defined all the operations of the Poisson brackets since an arbitrary function may be even or odd while H, like L, must be even. It leaves us free to define the Poisson brackets when a function in the position of H in Eq. (2.12) is odd. In viewing the quantization, the most important properties of the Poisson brackets are the algebraic ones, and so the Poisson brackets can be defined in such a way as to have an algebra over a Grassmann ring, that is,

$$\epsilon \{E, O\} = \{\epsilon E, O\} = \{E, \epsilon O\}, \tag{2.14}$$

where ϵ is an odd constant. With Eqs. (2.12), (2.13), and (2.14) we have

$$\{E_1, E_2\} = \left(\frac{\partial E_1}{\partial q_i}\frac{\partial E_2}{\partial p_i} - \frac{\partial E_2}{\partial q_i}\frac{\partial E_1}{\partial p_i}\right) + \left(\frac{\partial E_1}{\partial \xi_i}\frac{\partial E_2}{\partial \pi_i} - \frac{\partial E_2}{\partial \xi_i}\frac{\partial E_1}{\partial \pi_i}\right), \tag{2.15a}$$

$$\{O, E\} = -\{E, O\} = \left(\frac{\partial O}{\partial q_i}\frac{\partial E}{\partial p_i} - \frac{\partial E}{\partial q_i}\frac{\partial O}{\partial p_i}\right) - \left(\frac{\partial O}{\partial \xi_i}\frac{\partial E}{\partial \pi_i} + \frac{\partial E}{\partial \xi_i}\frac{\partial O}{\partial \pi_i}\right), \tag{2.15b}$$

$$\{O_1, O_2\} = \left(\frac{\partial O_1}{\partial q_i}\frac{\partial O_2}{\partial p_i} + \frac{\partial O_2}{\partial q_i}\frac{\partial O_1}{\partial p_i}\right) - \left(\frac{\partial O_1}{\partial \xi_i}\frac{\partial O_2}{\partial \pi_i} + \frac{\partial O_2}{\partial \xi_i}\frac{\partial O_1}{\partial \pi_i}\right), \tag{2.15c}$$

from which we obtain

$$\{\xi_i, \xi_j\} = \{\pi_i, \pi_j\} = 0, \tag{2.16a}$$

$$\{\xi_i, \pi_j\} = -\{\pi_j, \xi_i\} = -\delta_{ij}. \tag{2.16b}$$

Other algebraic properties of the Poisson brackets are listed in Ref. 24.

It is convenient to analyze the general algebraic structure of the Poisson brackets before quantization. Consider a set of homogeneous polynomials of degree k in Grassmann variables,

$$v_k = f(q_i, p_i)\xi_{i_1}\cdots\xi_{i_R}\pi_{j_1}\cdots\pi_{j_S} \quad (k = R - S), \tag{2.17}$$

where the degrees of the canonical variables are defined as

$$\deg(\xi_i) = +1, \quad \deg(\pi_i) = -1, \tag{2.18a}$$

$$\deg(q_i) = \deg(p_i) = 0. \tag{2.18b}$$

Obviously, v_k is an even or odd variable depending on whether k is even or odd. Thus we have

$$v_h v_k = (-1)^{hk} v_k v_h. \tag{2.19}$$

The properties of the Poisson brackets for the homogeneous elements are

$$\{v_h, v_k + w_k\} = \{v_h, v_k\} + \{v_h, w_k\}, \tag{2.20a}$$

$$\{v_h, v_k\} = -(-1)^{hk}\{v_k, v_h\}, \tag{2.20b}$$

$$\{v_l, v_m v_n\} = (-1)^{lm} v_m \{v_l, v_n\} + \{v_l, v_m\} v_n, \tag{2.20c}$$

$$\{v_l v_m, v_n\} = (-1)^{mn}\{v_l, v_n\} v_m + v_l \{v_m, v_n\}, \tag{2.20d}$$

$$(-1)^{ln}\{v_l, \{v_m, v_n\}\} + (-1)^{ml}\{v_m, \{v_n, v_l\}\} + (-1)^{nm}\{v_n, \{v_l, v_m\}\} = 0, \tag{2.20e}$$

which can be easily verified from the definitions given in Eqs. (2.15).

Now we want to define a quantum theory such that its limit $\hbar \to 0$ yields classical mechanics. Using the algebraic properties of the Poisson brackets of the Grassmann polynomials [Eqs. (2.20)] we can proceed as in Dirac's book.[25] Now let v_k be quantum variables (which are operators in a Hilbert space), and let the quantum Poisson brackets have the same properties as the classical ones in Eqs. (2.20), but not the properties (2.15)–(2.19) that give rise to them. We calculate the quantum mechanical Poisson brackets $\{v_l v_m, v_n v_p\}$ in two different ways, using Eqs. (2.20c) and (2.20d), respectively. By equating the results we get

$$\left[v_n v_l - (-1)^{nl} v_l v_n\right]\{v_m, v_p\} = \{v_n, v_l\}\left[v_m v_p - (-1)^{nl} v_p v_m\right] \tag{2.21}$$

from which the quantum mechanical Poisson brackets can be determined as

$$v_n v_l - (-1)^{nl} v_l v_n = c\{v_n, v_l\}, \tag{2.22}$$

with c a constant independent of v_l and v_n. If v contains no Grassmann variables, then c must be equal to $i\hbar$ from the usual quantization procedure. Thus the constant must be $i\hbar$ in the more general case. Therefore, the quantization rule is

$$v_n v_l - (-1)^{nl} v_l v_n = i\hbar\{v_n, v_l\}, \tag{2.23}$$

which can be specialized to the various cases of even and odd operators as

$$[E_1, E_2] = i\hbar\{E_1, E_2\}, \tag{2.24a}$$

$$[O, E] = i\hbar\{O, E\}, \tag{2.24b}$$

$$[O_1, O_2]_+ = i\hbar\{O_1, O_2\}. \tag{2.24c}$$

B. Classical spin-$\frac{1}{2}$ systems

It is well known that the quantum coherent states of Bose particles yield the amplitudes of classical oscillators in the limit $\hbar \to 0$. Analogously, Casalbuoni[24] showed that the quantum coherent states of Fermi particles yield in the classical limit ($\hbar \to 0$) the amplitudes of a system characterized by Grassmann (anticommuting) variables. From the propagator of the coherent-state representation of a quantum system consisting of both a boson and a fermion, he deduced the action of the system in the classical limit to be

$$\int L dt = \int \left[\frac{i}{2}\sum_{i=1}^{2}\xi_i\dot{\xi}_i + \frac{m}{2}\dot{q}^2 - V(q, \xi_i)\right] dt, \tag{2.25}$$

where ξ_i is a two-dimensional Grassmann variable and the bracketed quantity is the Lagrangian L.

Casalbuoni argued inductively that a fermion having internal degrees of freedom, such as spin, would lead to a similarly structured Lagrangian but involving an N-dimensional Grassmann variable, thus making the summation in Eq. (2.25) over N components. We call the summed term the kinetic Lagrangian; it is not a kinetic energy since it disappears from the Hamiltonian, it is first degree in the time derivative, and it is proportional to the imaginary unit. From the Euler-Lagrange equations we obtain the dynamic equations for the system as

$$m\ddot{q}_i = -\frac{\partial V}{\partial q_i}, \quad \dot{\xi}_i = -i\frac{\partial V}{\partial \xi_i}, \tag{2.26}$$

which shows that the dynamic equations for ξ_i are first-order differential equations with respect to time, just as the well-known torque equation for spins.

We now show using Dirac's procedure of quantization that a system with $N = 3$ corresponds to the classical nonrelativistic spin-$\frac{1}{2}$ system. From Eqs. (2.5) we obtain the canonical momenta from the Lagrangian in Eq. (2.25) (with N=3) to be

$$p_i = m\dot{q}_i, \quad \pi_i = -\frac{i}{2}\xi_i. \tag{2.27}$$

Note that unlike the usual canonical variables p_i and q_i the Grassmann canonical variables π_i and ξ_i are not linearly independent for the system described by the Lagrangian in Eq. (2.25). Therefore the expression for π_i is a constraint equation. By defining

$$\chi_i = \pi_i + \frac{i}{2}\xi_i, \tag{2.28}$$

we get from Eq. (2.16)

$$\{\chi_i, \chi_j\} = -i\delta_{ij}. \tag{2.29}$$

Hence, χ_i are second-class constraints.[25] From Eqs. (2.10) and (2.27) we also obtain the Hamiltonian of the system,

$$H = \frac{1}{2}\frac{p^2}{m} + V(q_i, \xi_i). \tag{2.30}$$

Note that the kinetic Lagrangian does not appear in the Hamiltonian, thus showing that it is not a kinetic energy. Also note that when second-class constraints exist in a system, the quantization procedure should use Dirac brackets[25] instead of Poisson brackets. For the homogeneous elements v_k defined in Eq. (2.17), the Dirac brackets are defined as

$$\{v_h, v_k\}^* = \{v_h, v_k\} - \{v_h, \chi_i\}\left(C^{-1}\right)_{ij}\{\chi_j, v_k\}, \tag{2.31}$$

where $\left(C^{-1}\right)_{ij}$ is defined through

$$C_{ij} = \{\chi_i, \chi_j\} = -i\delta_{ij}, \tag{2.32}$$

which leads to

$$\left(C^{-1}\right)_{ij} = i\delta_{ij}. \tag{2.33}$$

At this point it is straightforward to derive equations analogous to Eqs. (2.20) for the Dirac brackets. They in turn can be used to derive the analog of Eq. (2.23) in terms of Dirac brackets,

$$v_n v_l - (-1)^{nl} v_l v_n = i\hbar\{v_n, v_l\}^*. \tag{2.34}$$

Since no dependence on π_i appears in the Hamiltonian (2.30), we can assume that any physical quantity (E_1, E_2, E, O_1, O_2, and O) lacks that dependence. Their Dirac brackets, Eq. (2.31), thus simplify to

$$\{E_1, E_2\}^* = \left(\frac{\partial E_1}{\partial q_i}\frac{\partial E_2}{\partial p_i} - \frac{\partial E_2}{\partial q_i}\frac{\partial E_1}{\partial p_i}\right) + i\frac{\partial E_1}{\partial \xi_i}\frac{\partial E_2}{\partial \xi_i}, \tag{2.35a}$$

$$\{O, E\}^* = -\{E, O\}^* = \left(\frac{\partial O}{\partial q_i}\frac{\partial E}{\partial p_i} - \frac{\partial E}{\partial q_i}\frac{\partial O}{\partial p_i}\right) - i\frac{\partial O}{\partial \xi_i}\frac{\partial E}{\partial \xi_i}, \tag{2.35b}$$

$$\{O_1, O_2\}^* = \left(\frac{\partial O_1}{\partial q_i}\frac{\partial O_2}{\partial p_i} + \frac{\partial O_2}{\partial q_i}\frac{\partial O_1}{\partial p_i}\right) - i\frac{\partial O_1}{\partial \xi_i}\frac{\partial O_2}{\partial \xi_i}. \tag{2.35c}$$

By using Eqs. (2.35) we obtain

$$\{q_i, p_j\}^* = \delta_{ij}, \quad \{\xi_i, \xi_j\}^* = -i\delta_{ij}, \tag{2.36}$$

which give the fundamental quantization rules

$$[q_i, p_j] = i\hbar\delta_{ij}, \quad [\xi_i, \xi_j]_+ = \hbar\delta_{ij} \tag{2.37}$$

of the system according to Eq. (2.34). Obviously, the Grassmann variables become Fermi operators after quantization.

Also, by using Eqs. (2.35) we can obtain the Liouville equations

$$\dot{p}_i = \{p_i, H\}^* = -\frac{\partial V}{\partial q_i}, \tag{2.38a}$$

$$\dot{\xi}_i = \{\xi_i, H\}^* = -i\frac{\partial H}{\partial \xi_i} = -i\frac{\partial V}{\partial \xi_i}. \tag{2.38b}$$

With Eq. (2.27), the above equations are readily recognized as the Euler-Lagrange equations (2.26). This shows the correctness of Dirac's canonical formalism.

We have yet to determine the functional form of spin. It can be shown that if the infinitesimal canonical transformations are performed on the variables ξ_i only, then these transformations belong to a group O_N under which the ξ_i transform as the components of a N-component vector. For the three-component Grassmann vector ξ_i that is studied here, the transformation group is O_3 (homomorphic to SU_2), whose irreducible representation corresponds to Pauli matrices. From the quantization rule in Eq. (2.37) we obtain

$$\boldsymbol{\xi} = \sqrt{\hbar/2}\boldsymbol{\sigma}, \tag{2.39}$$

where $\boldsymbol{\sigma}$ is the vector of Pauli matrices. Therefore, we can find the classical spin-$\frac{1}{2}$ functions from the generators of the O_3 group which are

$$S_{ij} = -\frac{i}{2}[\xi_i, \xi_j] = -i\xi_i\xi_j = \epsilon_{ijl}S_l, \tag{2.40}$$

where $\mathbf{S}$ is a pseudovector defined as

$$\mathbf{S} = -\frac{i}{2}\boldsymbol{\xi}\times\boldsymbol{\xi}. \tag{2.41}$$

Note that anticommutativity prevents the vector product $\boldsymbol{\xi}\times\boldsymbol{\xi}$ from vanishing. (Note also for contrast that the scalar product $\boldsymbol{\xi}\cdot\boldsymbol{\xi}$ does vanish.)

If we calculate the Dirac brackets for $\mathbf{S}$, we obtain

$$\{S_i, S_j\}^* = \epsilon_{ijk}S_k. \tag{2.42}$$

Quantization of the above equation according to Eq. (2.34) (S_i are even variables) yields the operator equation

$$[S_i, S_j] = i\hbar\epsilon_{ijk}S_k, \tag{2.43}$$

which is the general quantum angular momentum commutator rule. By using Eqs. (2.39) and (2.42) the quantized $\mathbf{S}$ can be expressed as

$$\mathbf{S} = \frac{\hbar}{2}\boldsymbol{\sigma}. \tag{2.44}$$

Thus $\mathbf{S}$ is identified as the spin-$\frac{1}{2}$ operator, which proves that the classical pseudovector defined by Grassmann variables in Eq. (2.41) is indeed the spin vector.

Now let us consider the most general Lagrangian for a classical particle with spin moving in external fields. Since $\boldsymbol{\xi}$ in the potential energy can only appear quadratically, Eq. (2.25) (with $N = 3$) becomes

$$L = \frac{i}{2}\boldsymbol{\xi}\cdot\dot{\boldsymbol{\xi}} + \frac{1}{2}m\dot{q}^2 - V_1(\mathbf{q}) - \xi_i\xi_j V_{ij}(\mathbf{q}). \tag{2.45}$$

Thus, the tensor V_{ij} must be antisymmetric, that is,

$$V_{ij} = \frac{i}{2}\epsilon_{ijk}V_k(\mathbf{q}). \tag{2.46}$$

Hence we can rewrite the Lagrangian (2.45) as

$$L = \frac{i}{2}\boldsymbol{\xi}\cdot\dot{\boldsymbol{\xi}} + \frac{1}{2}m\dot{q}^2 - V_1(\mathbf{q}) + \mathbf{S}\cdot\mathbf{V}(\mathbf{q}). \tag{2.47}$$

By forming the Euler-Lagrange equations for ξ_i, we obtain

$$\dot{\boldsymbol{\xi}} = \boldsymbol{\xi}\times\mathbf{V}. \tag{2.48}$$

Using this equation we can obtain the dynamic equation for spin,

$$\dot{S}_i = -i\epsilon_{ijk}\xi_j\dot{\xi}_k = \epsilon_{ijk}S_jV_k \tag{2.49a}$$

or

$$\dot{\mathbf{S}} = \mathbf{S}\times\mathbf{V}. \tag{2.49b}$$

Thus the Lagrangian (2.47) yields the well-known torque equation for a classical spin.

It is important to realize that the kinetic Lagrangian term in Eq. (2.47) cannot be expressed in terms of $\mathbf{S}$ and $\dot{\mathbf{S}}$ alone. Thus anticommuting variables are needed at the Lagrangian level. However, the components of the classical spin function $\mathbf{S}$ commute, $[S_i, S_j] = 0$. Thus, at the equation of motion level of Eq. (2.49) a fully commuting theory exists.

Since the change in the spin vector is perpendicular to the spin vector in Eq. (2.49), its magnitude is a constant of the motion, a well-known fact. Note that no constraint was imposed in this theory on the classical Grassmann variable ξ to obtain this result. Note also that ξ represents the internal degrees of freedom of an individual particle in this theory. In contrast, previous theories of magnetism having continuum origins[13–17] introduced only the total saturated magnetization of the body and phenomenologically constrained its magnitude. The previous Lagrangian approach[18] used the magnetization of an individual particle as a Lagrangian variable and invoked a phenomenological constraint on its magnitude.

Finally, we note that the nonrelativistic Lagrangian theory for spin greater than $\frac{1}{2}$ has not been studied yet. However, based on the quantum theory of spin where there is no difference in formulation between spin $\frac{1}{2}$ and higher spin in a nonrelativistic case, we surmise that our formalism should accommodate the higher spin by altering the gyromagnetic ratio in the magnetic moment associated with the spin.

III. LAGRANGIAN DENSITY IN SPATIAL AND MATERIAL FRAMES

In this section, we extend the well-developed continuum Lagrangian formalism[2] to include spin magnetism by using Grassmann variables to describe the internal degrees of freedom of particles with spin in a dielectric crystal. We give only a summary derivation of the Lagrangian formalism here since a full exposition of the development is given in Ref. 2.

A. Total discrete Lagrangian

As mentioned earlier, our theory, though a macroscopic theory, is derived from microscopic physics by a long-wavelength limit. We regard a crystal as an array of point particles. We allow the primitive unit cell to contain N particles, which are labeled by a lowercase Greek letter, say, α. These particles should include all of the ions and, depending on the problem under study, one or two bonding electrons. Each particle has a fixed charge e^α and a fixed mass m^α. Its position is $\mathbf{x}^{\alpha n}$ where n, which has three integer components, labels the primitive unit cell of which the particle is a constituent. If the particle has spin, we attach the three-component Grassmann vector $\boldsymbol{\xi}^{\alpha n}$ to describe its internal (spin) degrees of freedom, just as the spatial vector $\mathbf{x}^{\alpha n}$ does to the mechanical degrees of freedom.

The particles reside in a vacuum and are subject to bonding forces between themselves. The bonding forces are *short-range* forces and are described by a potential energy $V(\mathbf{x}^{\alpha n}, \boldsymbol{\xi}^{\alpha n})$. As we see later, the generalized expansion of this potential produces all the possible bonding force energy terms including the exchange energy of particles with spin derived by Akhiezer *et al.*[26] There are also macroscopic (or *long-range*) electric and magnetic fields existing in the vacuum around the particle that give rise to body forces. These electric and magnetic fields can be created by the medium itself or can be created externally. Their interaction energy with the medium is included in the interaction Lagrangian.

The total Lagrangian L of this system consists of three parts

$$L = L_M + L_I + L_F, \tag{3.1}$$

the matter Lagrangian L_M, the electromagnetic field Lagrangian L_F, and the field-matter interaction Lagrangian L_I. They are defined through

$$L_M = \frac{1}{2}\sum_{\alpha,n}\left(m^\alpha(\dot{\mathbf{x}}^{\alpha n})^2 + i\boldsymbol{\xi}^{\alpha n}\cdot\dot{\boldsymbol{\xi}}^{\alpha n}\right) - V(\mathbf{x}^{\alpha n},\boldsymbol{\xi}^{\alpha n}), \quad (3.2)$$

$$L_F = \int \frac{1}{2}\left(\epsilon_0\mathbf{E}^2 - \frac{1}{\mu_0}\mathbf{B}^2\right)dv, \quad (3.3)$$

$$L_I = \sum_{\alpha,n}\{e^\alpha[\dot{\mathbf{x}}^{\alpha n}(t)\cdot\mathbf{A}(\mathbf{x}^{\alpha n},t) - \Phi(\mathbf{x}^{\alpha n},t)] + \mu^\alpha\mathbf{s}^{\alpha n}(\mathbf{x}^{\alpha n},t)\cdot\mathbf{B}(\mathbf{x}^{\alpha n},t)\}, \quad (3.4)$$

where $dv \equiv dz_1dz_2dz_3$ is the spatial coordinate system volume element. In the above equation, $\mathbf{E}$ and $\mathbf{B}$ are electric and magnetic induction fields expressed as functions of the scalar and vector potentials,

$$\mathbf{E} = -\boldsymbol{\nabla}\Phi - \frac{\partial\mathbf{A}}{\partial t}, \quad (3.5)$$

$$\mathbf{B} = \boldsymbol{\nabla}\times\mathbf{A}, \quad (3.6)$$

and

$$\mathbf{s}^{\alpha n} = -\frac{i}{2}\boldsymbol{\xi}^{\alpha n}\times\boldsymbol{\xi}^{\alpha n} \quad (3.7)$$

is the spin of the α-particle in the n primitive cell. $\mu^\alpha \equiv g_s e^\alpha/2m^\alpha$ is the magnetic moment arising from the spin of the α-particle and g_s is the gyromagnetic ratio. By choosing the interaction energy of spin magnetization with the magnetic induction field in Eq. (3.4) to have the same form as that in the well-known nonrelativistic quantum theory of spin, we guarantee that the quantization of our formalism corresponds to the correct quantum theory of spin $\frac{1}{2}$. Also from this point of view, we surmise that our formalism can be extended to include higher spin by altering g_s since there is no difference in the forms of spin interaction energy for different spin values in nonrelativistic quantum theory.

B. Continuum limit

The continuum limit of the discrete Lagrangian can be done by replacing the discrete cell index n (which contains three components) by a continuous variable $\mathbf{X}$, that is, $n \to \mathbf{X}$. This replacement, by not affecting the index α, retains all modes of motion of the various sublattices and all the symmetry and anisotropy of the crystal. It also fulfills the same function of labeling the matter as the discrete index n did. Therefore we call $\mathbf{X}$ the continuum material coordinate vector. All the mechanical and internal coordinates can be replaced by their continuum counterparts:

$$\mathbf{x}^{\alpha n}(t) \to \mathbf{x}^\alpha(\mathbf{X},t), \quad \boldsymbol{\xi}^{\alpha n}(t) \to \boldsymbol{\xi}^{T\alpha}(\mathbf{X},t)\,\Omega_0^{\frac{1}{2}}, \quad (3.8)$$

where T stands for total, meaning it consists of a spontaneous value, and a variation from that value, and Ω_0 is the volume of a primitive unit cell. Also in the continuum limit sums over the cell index n become integrals over the continuous material coordinate $\mathbf{X}$, that is,

$$\sum_n F(\mathbf{x}^{\alpha n}(t),\boldsymbol{\xi}^{\alpha n}(t)) \to \frac{1}{\Omega_0}\int F(\mathbf{x}^\alpha(\mathbf{X},t),\boldsymbol{\xi}^{T\alpha}(\mathbf{X},t))dV, \quad (3.9)$$

where $dV \equiv dX_1dX_2dX_3$. We can also define the mass density and charge density as

$$\rho^\alpha \equiv m^\alpha/\Omega_0, \quad q^\alpha \equiv e^\alpha/\Omega_0. \quad (3.10)$$

C. Center-of-mass and internal motion fields

It is easy to see that the continuum measure of the center-of-mass position of a primitive unit cell,

$$\mathbf{x}(\mathbf{X},t) = \frac{\sum_\alpha \rho^\alpha\mathbf{x}^\alpha(\mathbf{X},t)}{\sum_\alpha \rho^\alpha}, \quad (3.11)$$

represents the position vector for a mass point in continuum mechanics. Since the material coordinate $\mathbf{X}$ represents the undeformed position of the mass point that it labels, the spatial (or deformed) position $\mathbf{x}$ as a function of $\mathbf{X}$ and t, $\mathbf{x} = \mathbf{x}(\mathbf{X},t)$, or its inverse, $\mathbf{X} = \mathbf{X}(\mathbf{x},t)$, is called the deformation transformation. The difference between the deformed and undeformed positions of a mass point is defined as the displacement vector

$$u_i = x_i - \delta_{iA}X_A. \quad (3.12)$$

Note that spatial frame components are denoted by lowercase subscripts and material frame components are denoted by uppercase subscripts even though the choice of the Kronecker δ as the shifter tensor in the Eq. (3.12) shows that both types of coordinates are referred to the same rectangular Cartesian axes. The displacement vector $\mathbf{u}$ (or equivalently $\mathbf{x}$) carries all the momentum of the material medium. Its dynamic equation describes the three acoustic modes.

The other linear combinations of coordinates describe $3N-3$ internal degrees of vibration and are called internal coordinates or internal motion fields. They are defined by

$$\mathbf{y}^{T\mu} = \sum_{\alpha=1}^{N} U^{\mu\alpha}\mathbf{x}^\alpha(\mathbf{X},t), \quad \mu = 1,2,\ldots,N-1, \quad (3.13)$$

where T has the same meaning as used in $\boldsymbol{\xi}^{T\alpha}$. Since all the momentum of the material medium is carried by the center-of-mass field, the other linear combinations of coordinates must not carry any momentum of the material medium. This can be guaranteed by requiring the internal coordinates to be invariant to displacements of the crystal in space, that is,

$$\sum_{\alpha=1}^{N} U^{\mu\alpha} = 0. \quad (3.14)$$

If we let $\mathbf{y}^{T0}$ be $\mathbf{x}$, we obtain $U^{0\alpha} = \rho^\alpha/\rho^0$ where $\rho^0 = \sum_\alpha \rho^\alpha$. We denote the inverse transformation as

$$\mathbf{x}^\alpha = \sum_{\mu=0}^{N-1} V^{\alpha\mu}\mathbf{y}^{T\mu}, \quad \alpha = 1,2,\ldots,N, \tag{3.15}$$

with orthogonality relations

$$\sum_{\alpha=1}^{N} U^{\mu\alpha}V^{\alpha\nu} = \delta^{\mu\nu}, \quad \sum_{\nu=0}^{N-1} V^{\alpha\nu}U^{\nu\beta} = \delta^{\alpha\beta}. \tag{3.16}$$

We can also choose the new coordinates so that the kinetic energy is diagonal for further convenience, that is,

$$\sum_{\alpha=1}^{N} \rho^\alpha \left(\dot{\mathbf{x}}^\alpha\right)^2 = \sum_{\nu=0}^{N-1} m^\nu \left(\dot{\mathbf{y}}^{T\nu}\right)^2, \tag{3.17}$$

from which we obtain

$$m^\nu U^{\nu\alpha} = \rho^\alpha V^{\alpha\nu}. \tag{3.18}$$

The Grassmann vector $\boldsymbol{\xi}^\alpha$ describes the internal degrees of freedom of a particle with spin. Thus, it does not carry any momentum of the medium.

D. Matter Lagrangian density and stored energy

As seen previously, the matter Lagrangian is the difference between the sum of the kinetic energy of motion and the kinetic Lagrangian of spin and the potential or stored energy that describes all the short-range interactions of the material medium. The matter Lagrangian density is defined in the two reference frames through

$$L_M = \int \mathcal{L}_{MM} dV = \int \mathcal{L}_{MS} dv. \tag{3.19}$$

It can be obtained most naturally in the material frame from Eq. (3.2),

$$\mathcal{L}_{MM} = \frac{1}{2}\rho^0\left(\dot{\mathbf{x}}\right)^2 + \frac{1}{2}\sum_{\nu=1}^{N-1} m^\nu \left(\dot{\mathbf{y}}^{T\nu}\right)^2 + \frac{i}{2}\sum_{\alpha=1}^{N} \boldsymbol{\xi}^{T\alpha}\cdot\dot{\boldsymbol{\xi}}^{T\alpha} - \rho^0\Sigma(\mathbf{y}^{T\nu}(\mathbf{X},t), \mathbf{y}^{T\nu}_{,A}(\mathbf{X},t), \boldsymbol{\xi}^{T\alpha}(\mathbf{X},t), \boldsymbol{\xi}^{T\alpha}_{,A}(\mathbf{X},t)), \tag{3.20}$$

where $\rho^0\Sigma$ is the stored energy density in the continuum limit.[2] Since the volume elements $dv = J(\mathbf{x}/\mathbf{X})\,dV$ are related by the Jacobian between spatial and material frames, the matter Lagrangian density in the spatial frame in Eq. (3.19) can be obtained from that in the material frame by multiplying by J^{-1}. This also gives the spatial frame or deformed mass density as $\rho = \rho^0/J$.

As seen from the previous section, the Hamiltonian of a nonrelativistic classical spinning particle is identical to its quantized one. We know that the Hamiltonian of a nonrelativistic quantum mechanical model of spin contains only spin operators. Therefore, we conclude that the stored energy contains only spin functions instead of other functions of Grassmann variables.

The stored energy is constructed as general as possible while conserving energy, momentum, and angular momentum. These require, in order, that the stored energy must not be an explicit function of time, $\mathbf{z}$ or $\mathbf{x}$, and be a function of a minimal but complete set of rotationally invariant measures of the mechanical coordinates and the spin density vectors and their first derivatives. Since perturbations of the medium are typically small, a series expansion in terms of the rotational invariants can be truncated at a finite number of terms for some particular interaction only if the rotational invariants vanish in the natural state of the crystal. These several requirements are met with the choice of

$$\Pi^\nu_A = X_{A,i}\left(\delta_{iB}Y^\nu_B + y^\nu_i\right) - Y^\nu_A, \tag{3.21a}$$
$$\Pi^\nu_{AB} = X_{A,i}y^\nu_{i,B}, \tag{3.21b}$$
$$\Gamma^\alpha_A = X_{A,i}\left(\delta_{iB}S^\alpha_B + s^\alpha_i\right) - S^\alpha_A, \tag{3.21c}$$
$$\Gamma^\alpha_{AB} = X_{A,i}s^\alpha_{i,B}, \tag{3.21d}$$
$$E_{AB} = \left(x_{i,A}x_{i,B} - \delta_{AB}\right)/2, \tag{3.21e}$$

as rotational invariants, where

$$y^{T\nu}_i = \delta_{iB}Y^\nu_B + y^\nu_i, \tag{3.22a}$$
$$s^{T\alpha}_i = -\frac{i}{2}\epsilon_{ijk}\xi^{T\alpha}_j\xi^{T\alpha}_k = \delta_{iB}S^\alpha_B + s^\alpha_i \tag{3.22b}$$

are used. Y^ν_B and S^α_B are natural-state or spontaneous values of $y^{T\nu}_i$ and $s^{T\alpha}_i$. y^ν_i and s^α_i represent deviations from the natural state. The stored energy can now be written as

$$\rho^0\Sigma = J\left(\mathbf{x}/\mathbf{X}\right)\rho\Sigma\left(\Pi^\nu_A, \Pi^\nu_{AB}, \Gamma^\alpha_A, \Gamma^\alpha_{AB}, E_{AB}\right). \tag{3.23}$$

We now look at some typical terms of the rotational invariants of the spin functions and their derivatives in the expansion of the stored energy. In order to compare with those obtained by Akhiezer *et al.*,[26] we consider a purely optical phenomenon at frequencies to which acoustic modes cannot respond. Therefore, we can set all coordinates expressing deformation to zero, which is equivalent to setting u_i to zero in Eq. (3.12). Thus the difference between spatial and material coordinates disappears simplifying the rotational invariants to $\Gamma^\alpha_i = s^\alpha_i$ and $\Gamma^\alpha_{ij} = s^\alpha_{i,j}$.

If we compare the terms in the series expansion of the stored energy with those obtained by Akhiezer *et al.*[26] we can see that the quadratic term of Γ^α_{ij} corresponds to the exchange energy between particles with spin, which is the most important short-range force potential for spin in ferromagnetic and antiferromagnetic dielectric crystals. Also in these materials, spins are oriented on each sublattice in the natural state, which means S^α_i are not all zero. Therefore the term linear in Γ^α_i in the series expansion of $\rho^0\Sigma$ is important, just as the spontaneous polarization in a ferroelectric material. It corresponds to the spin magnetic dipole interaction energy in a mean field that is generated by the spins themselves. When combined with the interaction Lagrangian, we can interpret its relation with the spontaneous magnetic induction field in the medium. We discuss this further in the next section in which we study the equation of motion for each excitation mode. The quadratic term in Γ_i^α can be considered as a higher-order contribution of the same interaction. The term linear in Γ_{jk}^α is a part of the exchange energy, which exists only if the crystal lacks a center of symmetry, and the bilinear term of Γ_i^α and Γ_{jk}^α can be considered as its higher-order term. According to Akhiezer *et al.*[26] both of these two terms are usually negligible.

E. Interaction Lagrangian and multipole expansion

The continuum limit of the interaction Lagrangian of Eq. (3.4) is

$$L_I = \int \mathcal{L}_{IM} dV = \int \mathcal{L}_{IS} dv, \quad (3.24)$$

$$\mathcal{L}_{IS} = J^{-1}\mathcal{L}_{IM}, \quad (3.25)$$

where the spatial frame interaction Lagrangian density that is not related to spin is

$$\mathcal{L}_{IS}^c = \mathbf{j}^c \cdot \mathbf{A} - q\Phi \quad (3.26)$$

and

$$q(\mathbf{z},t) = \sum_\alpha q^\alpha \int \delta(\mathbf{z} - \mathbf{x}^\alpha(\mathbf{X},t))dV, \quad (3.27)$$

$$\mathbf{j}^c(\mathbf{z},t) = \sum_\alpha q^\alpha \int \dot{\mathbf{x}}^\alpha(\mathbf{X},t)\,\delta(\mathbf{z} - \mathbf{x}^\alpha(\mathbf{X},t))dV. \quad (3.28)$$

The charge and current densities can be expanded in terms of $\mathbf{u}^\alpha \equiv \mathbf{x}^\alpha - \mathbf{x}$ since all particles in a unit cell are localized in dielectrics and thus the expansion is guaranteed to converge in the long-wavelength region. By invoking the dielectric assumption or charge neutrality of a unit cell, $\sum_\alpha q^\alpha = 0$, multipole expansions of Eqs. (3.27) and (3.28) yield the dielectric charge and current densities[2]

$$q^D(\mathbf{z},t) = -\nabla\cdot\mathbf{P} + \nabla\nabla : \overleftrightarrow{Q}, \quad (3.29)$$

$$\mathbf{j}^D(\mathbf{z},t) = \frac{\partial\mathbf{P}}{\partial t} + \nabla\times(\mathbf{P}\times\dot{\mathbf{x}}) - \frac{\partial}{\partial t}\left(\nabla\cdot\overleftrightarrow{Q}\right) - \nabla\times\left[\nabla\cdot\left(\overleftrightarrow{Q}\times\dot{\mathbf{x}}\right)\right] + \nabla\times\mathbf{M}^c, \quad (3.30)$$

where the polarization $\mathbf{P}$, quadrupolarization $\overleftrightarrow{Q}$, and magnetization $\mathbf{M}^c$ from the motion of bound charge are given by

$$\mathbf{P} = \sum_\alpha q^\alpha \frac{\mathbf{u}^\alpha}{J} = \sum_\nu \frac{q^\nu \mathbf{y}^{T\nu}}{J}, \quad (3.31)$$

$$\overleftrightarrow{Q} = \frac{1}{2}\sum_\alpha q^\alpha \frac{\mathbf{u}^\alpha\mathbf{u}^\alpha}{J} = \frac{1}{2}\sum_{\mu\nu} \frac{q^{\mu\nu}\mathbf{y}^{T\mu}\mathbf{y}^{T\nu}}{J}, \quad (3.32)$$

$$\mathbf{M}^c = \frac{1}{2}\sum_\alpha \frac{q^\alpha \mathbf{u}^\alpha\times\dot{\mathbf{u}}^\alpha}{J} = \frac{1}{2}\sum_{\mu\nu} \frac{q^{\mu\nu}\mathbf{y}^{T\mu}\times\dot{\mathbf{y}}^{T\nu}}{J}. \quad (3.33)$$

Here q^ν, $q^{\mu\nu}$ are charge densities defined as

$$q^\nu \equiv \sum_\alpha q^\alpha V^{\alpha\nu}, \quad q^{\mu\nu} \equiv \sum_\alpha q^\alpha V^{\alpha\mu}V^{\alpha\nu}. \quad (3.34)$$

When the range of summation over internal coordinate designations μ, ν is not shown, a range $1, 2, \ldots, N-1$ is implied.

We treat the spin interaction with a magnetic field similarly. From Eq. (3.4) we know

$$\mathcal{L}_{IS}^s = \sum_\alpha \int \delta(\mathbf{z} - \mathbf{x}^\alpha(\mathbf{X},t)) \times \mu^\alpha \mathbf{s}^{T\alpha}(\mathbf{x}^\alpha(\mathbf{X},t),t)\cdot\mathbf{B}(\mathbf{z},t)\,dV. \quad (3.35)$$

Therefore we can define the spin magnetization as

$$\mathbf{M}^s(\mathbf{z},t) = \sum_\alpha \mu^\alpha \int \mathbf{s}^{T\alpha}(\mathbf{x}^\alpha(\mathbf{X},t),t)\delta(\mathbf{z} - \mathbf{x}^\alpha(\mathbf{X},t))dV, \quad (3.36)$$

thus giving

$$\mathcal{L}_{IS}^s = \mathbf{M}^s\cdot\mathbf{B}. \quad (3.37)$$

We carry out the multipole expansion in terms of $\mathbf{u}^\alpha$ as follows:

$$\begin{aligned}\mathbf{M}^s &= \sum_\alpha \mu^\alpha \int \mathbf{s}^{T\alpha}(\mathbf{x}+\mathbf{u}^\alpha)\,\delta(\mathbf{z}-\mathbf{x}-\mathbf{u}^\alpha)\,J^{-1}dv \\ &= \sum_\alpha \mu^\alpha \int \left(\mathbf{s}^{T\alpha} + \mathbf{u}^\alpha\cdot\nabla|_\mathbf{x}\, s^{T\alpha}\right)[\delta(\mathbf{z}-\mathbf{x}) \\ &\quad -\mathbf{u}^\alpha\cdot\nabla|_\mathbf{z}\,\delta(\mathbf{z}-\mathbf{x})]\,J^{-1}dv \\ &= \sum_\alpha J^{-1}\mu^\alpha\mathbf{s}^{T\alpha} - \sum_\alpha \mu^\alpha\left[\nabla\cdot\left(\mathbf{u}^\alpha J^{-1}\right)\right]\mathbf{s}^{T\alpha}. \end{aligned} \quad (3.38)$$

Since we only keep magnetization at the dipole level, the last term in the above equation can be dropped. Therefore we have

$$\mathbf{M}^s = \sum_\alpha J^{-1}\mu^\alpha\mathbf{s}^{T\alpha}(\mathbf{z},t). \quad (3.39)$$

We can define a spin current $\mathbf{j}^s$ so that we can rewrite $\mathcal{L}_{IS}^s$ as

$$\mathcal{L}_{IS}^s = \mathbf{j}^s\cdot\mathbf{A}. \quad (3.40)$$

This can be done by rearranging the term (3.37) into a perfect derivative minus a new term. The perfect derivative can be dropped as being unable to affect the equations of motion, which then yields

$$\mathbf{j}^s = \boldsymbol{\nabla} \times \mathbf{M}^s. \tag{3.41}$$

Now we can write the total $\mathcal{L}_{IS}$ by using the multipole expansion as

$$\mathcal{L}_{IS} = \mathbf{j} \cdot \mathbf{A} - q^D \Phi, \tag{3.42}$$

where

$$\mathbf{j} \equiv \mathbf{j}^D + \mathbf{j}^s \tag{3.43}$$

is the total bound current.

Although the interaction Lagrangian density of Eq. (3.42) can be used with charge and current densities, there is an interpretive value to transforming $\mathcal{L}_{IS}$ to a function of the fields $\mathbf{E}$ and $\mathbf{B}$, which, of course, are still functions of the Lagrangian variables $\mathbf{A}$ and Φ. This is done similarly to the change from Eq. (3.40) to Eq. (3.41). The result is

$$\begin{aligned}\mathcal{L}_{IS} = {} & \mathbf{P} \cdot (\mathbf{E} + \dot{\mathbf{x}} \times \mathbf{B}) - \mathbf{E} \cdot \boldsymbol{\nabla} \cdot \overleftrightarrow{Q} \\ & -\mathbf{B} \cdot \left[\left(\boldsymbol{\nabla} \cdot \overleftrightarrow{Q} + \overleftrightarrow{Q} \cdot \boldsymbol{\nabla}\right) \times \dot{\mathbf{x}}\right] + \mathbf{M} \cdot \mathbf{B},\end{aligned} \tag{3.44}$$

where $\mathbf{M} \equiv \mathbf{M}^c + \mathbf{M}^s$ is the total magnetization.

F. Field Lagrangian density and total Lagrangian density

From Eq. (3.3) the spatial frame field Lagrangian density is

$$\mathcal{L}_{FS} = \frac{1}{2}\left(\epsilon_0 \mathbf{E}^2 - \frac{1}{\mu_0}\mathbf{B}^2\right) \tag{3.45}$$

(the corresponding material frame density is not needed). The total Lagrangian density is simply the sum of the matter Lagrangian density, interaction Lagrangian density, and field Lagrangian density,

$$\mathcal{L} = \mathcal{L}_M + \mathcal{L}_I + \mathcal{L}_F. \tag{3.46}$$

Construction of the Lagrangian density is now complete. In the next section, we give the resulting dynamic equations for all the excitation modes in ferromagnetic, antiferromagnetic, and ferrimagnetic dielectrics.

IV. DYNAMICAL EQUATIONS

A. Maxwell-Lorentz equations

The Lagrangian density developed in the preceding section is a function of the Lagrangian variables $\mathbf{x}$, $\mathbf{y}^{T\nu}$ ($\nu = 1, 2, \ldots, N-1$), $\xi^{T\alpha}$ ($\alpha \leq N$), Φ, and $\mathbf{A}$, and an equation of motion for each can be found. The Maxwell-Lorentz equations can be obtained most naturally in the spatial frame from Euler-Lagrange equations for Φ and $\mathbf{A}$. This is because the field and interaction Lagrangian densities (3.42) and (3.45) are most easily expressed in the spatial frame. The Euler-Lagrange equation for Φ readily yields

$$\epsilon_0 \nabla \cdot \mathbf{E} = q^D, \tag{4.1}$$

where q^D is the dielectric charge density given by Eq. (3.29). Similarly the Euler-Lagrange equation for $\mathbf{A}$ yields

$$\frac{1}{\mu_0}\nabla \times \mathbf{B} - \epsilon_0 \frac{\partial \mathbf{E}}{\partial t} = \mathbf{j}, \tag{4.2}$$

where $\mathbf{j}$ is the total bound current consisting of dielectric current and spin current given in Eq. (3.43).

The electromagnetic equations (4.1) and (4.2) are in the Maxwell-Lorentz form in which $\mathbf{E}$ and $\mathbf{B}$ are regarded as the fundamental electromagnetic fields and the response of the matter is determined entirely by q^D and $\mathbf{j}$ which are expressed in terms of the matter response fields $\mathbf{P}$, $\mathbf{M}^c$, $\overleftrightarrow{Q}$, and $\mathbf{M}^s$. They are functions of mechanical coordinates and spin densities of the matter defined by Eqs. (3.31), (3.32), (3.33), and (3.39) in our Lagrangian formalism.

The Maxwell-Lorentz equations in conjunction with Eqs. (3.29), (3.30), and (3.41) can be reexpressed in the Maxwell form by defining the electric displacement field $\mathbf{D}$ and the magnetic field $\mathbf{H}$ through

$$\mathbf{D} \equiv \epsilon_0 \mathbf{E} + \mathbf{P} - \nabla \cdot \overleftrightarrow{Q}, \tag{4.3}$$

$$\mathbf{H} \equiv \mathbf{B}/\mu_0 - \mathbf{P} \times \dot{\mathbf{x}} - \mathbf{M} + \nabla \cdot \left(\overleftrightarrow{Q} \times \dot{\mathbf{x}}\right). \tag{4.4}$$

Thus, the Maxwell equations for a dielectric are

$$\nabla \cdot \mathbf{D} = 0, \tag{4.5}$$

$$\nabla \times \mathbf{H} - \partial \mathbf{D}/\partial t = 0, \tag{4.6}$$

$$\nabla \times \mathbf{E} + \partial \mathbf{B}/\partial t = 0, \tag{4.7}$$

$$\nabla \cdot \mathbf{B} = 0, \tag{4.8}$$

where the last two equations are the direct consequences of the definitions of $\mathbf{E}$ and $\mathbf{B}$ in terms of vector and scalar potentials given by Eqs. (3.5) and (3.6).

B. Dynamic equations for internal motion fields

The Euler-Lagrange equations for internal motion fields in the material frame yield

$$\begin{aligned} m^\nu \ddot{y}_i^{T\nu} - \frac{1}{2}\sum_\mu q^{\mu\nu}\epsilon_{ijk}\dot{y}_j^{T\mu}B_k - \frac{1}{2}\sum_\mu q^{\mu\nu}\epsilon_{ijk}y_j^{T\mu}\dot{B}_k = {} & q^\nu\left(E_i + \epsilon_{ijk}\dot{x}_j B_k\right) + \frac{1}{2}\sum_\mu q^{\mu\nu}y_j^{T\mu}\left(E_{j,i} + \epsilon_{jkl}\dot{x}_k B_{l,i}\right) \\ & + \frac{1}{2}\sum_\mu q^{\mu\nu}y_j^{T\mu}\left(E_{i,j} + \epsilon_{ikl}\dot{x}_k B_{l,j}\right) \\ & + \frac{1}{2}\sum_\mu q^{\mu\nu}\epsilon_{ijk}\dot{y}_j^{T\mu}B_k - \frac{\partial \rho_0 \Sigma}{\partial y_i^{T\nu}} + \frac{d}{dX_A}\frac{\partial \rho_0 \Sigma}{\partial y_{i,A}^{T\nu}}. \end{aligned} \tag{4.9}$$

This equation can be simplified by expanding the total time derivative of the magnetic induction field $\dot{B}_k$ and using two of the Maxwell equations, Eqs. (4.7) and (4.8), to obtain the dynamic equations for $\mathbf{y}^{T\mu}$,

$$m^\nu \ddot{y}_i^{T\nu} = q^\nu \mathcal{E}_i + \sum_\mu q^{\mu\nu} y_j^{T\mu} \mathcal{E}_{i,j} + \sum_\mu q^{\mu\nu} \epsilon_{ijk} \dot{y}_j^{T\mu} B_k - \sum_\mu q^{\mu\nu} \epsilon_{ikl} y_j^{T\mu} \dot{x}_{k,j} B_l - \frac{\partial \rho_0 \Sigma}{\partial y_i^{T\nu}} + \frac{d}{dX_A} \frac{\partial \rho_0 \Sigma}{\partial y_{i,A}^{T\nu}}, \tag{4.10}$$

where

$$\mathcal{E} \equiv \mathbf{E} + \dot{\mathbf{x}} \times \mathbf{B}. \tag{4.11}$$

C. Dynamic equation for spin

The Euler-Lagrange equations for the Grassmann variables $\boldsymbol{\xi}^{T\alpha}$ yield in the material frame

$$\dot{\xi}_i^{T\alpha} = \epsilon_{ijk} \xi_j^{T\alpha} \left(\mu^\alpha B_k - \frac{\partial \rho^0 \Sigma}{\partial s_k^{T\alpha}} + \frac{d}{dX_A} \frac{\partial \rho^0 \Sigma}{\partial s_{k,A}^{T\alpha}} \right). \tag{4.12}$$

Similarly to the manipulations in Sec. II, we obtain the dynamic equations for the spin density from Eqs. (3.22b) and (4.12) with the well-known torque equation

$$\frac{d\mathbf{s}^{T\alpha}}{dt} = \mu^\alpha \mathbf{s}^{T\alpha} \times {}^{\text{eff}}\mathbf{B}^\alpha \tag{4.13}$$

resulting, where ${}^{\text{eff}}\mathbf{B}^\alpha$ is an effective magnetic induction field exerted on the spin of the α-sublattice expressed as

$${}^{\text{eff}}B_k^\alpha = B_k - \frac{1}{\mu^\alpha} \left(\frac{\partial \rho^0 \Sigma}{\partial s_k^{T\alpha}} - \frac{d}{dX_A} \frac{\partial \rho^0 \Sigma}{\partial s_{k,A}^{T\alpha}} \right). \tag{4.14}$$

In ferromagnetic, antiferromagnetic, or ferrimagnetic materials, the spins in each sublattice can be aligned along one or more directions in the crystal; that is, the spontaneous or natural-state values of sublattice spin $\mathbf{S}^\alpha$ are not zero. In order to simplify the following discussion, we assume that the geometric shape of the crystal guarantees that the spontaneous magnetic field is homogeneous in the entire crystal. If we expand the stored energy based on the rotational invariants in Eqs. (3.21) to the linear term in $\mathbf{s}^{T\alpha}$ and setting $\mathbf{s}^\alpha$ to zero, we obtain

$$\mathbf{S}^\alpha \times {}^{\text{eff}}\mathbf{B}^{S\alpha} = 0, \tag{4.15}$$

where the spontaneous value of the effective magnetic induction field is defined as

$${}^{\text{eff}}B_A^{S\alpha} = B_A^S - \frac{M_A^\alpha}{\mu^\alpha} \tag{4.16}$$

and M_A^α is the coefficient of Γ_A^α in the stored energy expansion. Generally speaking, Eq. (4.15) is satisfied if $\mathbf{S}^\alpha || {}^{\text{eff}}\mathbf{B}^{S\alpha}$ is satisfied. However, the interaction energy density of spin magnetization with the magnetic induction field is a part of the total energy, just as the stored energy is. If we use the fact that the natural state of matter is the minimum free energy state and $\mathbf{B}^S$ is known, ${}^{\text{eff}}\mathbf{B}^{S\alpha} = 0$ should be satisfied and thus can be used to determine the coefficients of the linear terms of $\mathbf{s}^\alpha$ in the stored energy expansion. An analogous example that shows the relation between spontaneous electric field and spontaneous polarization in a ferroelectric dielectric is given in Chap. 8 of Ref. 2.

D. Dynamic equation for the center-of-mass field

The dynamic equation for the center-of-mass field can be obtained from the Euler-Lagrange equation for $\mathbf{x}$ in the material frame to be

$$\rho^0 \ddot{x}_i = -\epsilon_{ijk} B_j \dot{p}_k + \epsilon_{ijk} B_{j,l} \dot{q}_{kl} + \epsilon_{ijk} \dot{x}_j B_{k,l} p_l + \epsilon_{ijk} \dot{x}_j B_{k,lm} q_{lm} + E_{i,k} p_k + E_{i,jl} q_{jl} + m_j B_{j,i} + \frac{d}{dX_A} \frac{\partial \rho^0 \Sigma}{\partial x_{i,A}}, \tag{4.17}$$

where $\mathbf{p} \equiv J\mathbf{P}$, $\overleftrightarrow{q} \equiv J\overleftrightarrow{Q}$, and $\mathbf{m} \equiv J\mathbf{M}$ are polarization, quadrupolarization, and total magnetization in the material frame. A spatial frame form of Eq. (4.17) useful for deriving the momentum conservation law in the next section can be found by the manipulations described before[2] to be

$$\rho \ddot{x}_i = qE_i + \epsilon_{ijk} j_j B_k + t_{il,l}^E, \tag{4.18}$$

where t_{il}^E is the elastic stress tensor defined as

$$t_{il}^E = \mathcal{E}_i P_l + \epsilon_{ijk} \left(\frac{\partial Q_{lj}}{\partial t} + (Q_{lj} \dot{x}_m)_{,m} + \epsilon_{ljm} M_m \right) B_k - 2\epsilon_{ijk} Q_{lm} \dot{x}_{j,m} B_k + 2Q_{lm} \mathcal{E}_{i,m} - (Q_{lm} \mathcal{E}_i)_{,m} + J^{-1} \frac{\partial \rho^0 \Sigma}{\partial x_{i,A}} x_{l,A}. \tag{4.19}$$

V. CONSERVATION LAWS

A. Spin conservation

The form of the spin precession equation (4.13) leads immediately to the spin conservation law. By forming a scalar product of Eq. (4.13) with $J^{-1}\mathbf{s}^{T\alpha}$, we obtain

$$\frac{1}{J} \frac{d}{dt} \left(\mathbf{s}^{T\alpha} \cdot \mathbf{s}^{T\alpha} \right) = \frac{\partial}{\partial t} \left(\frac{\mathbf{s}^{T\alpha} \cdot \mathbf{s}^{T\alpha}}{J} \right) + \frac{\partial}{\partial z_l} \left(\frac{\mathbf{s}^{T\alpha} \cdot \mathbf{s}^{T\alpha} \dot{x}_l}{J} \right) = 0, \tag{5.1}$$

which states that the magnitude of each sublattice spin is conserved. The expression on the left of Eq. (5.1) is the material frame form of the law while that on the right is the spatial frame form.

B. Momentum conservation

Since momentum is inherently a spatial frame quantity, it is natural to express its conservation law in the spatial description, that is, with $\mathbf{z}, t$ as the independent variables. First we obtain a momentum continuity equation from the equation of motion of the center-of-mass field that carries all the momentum of the matter. Equation (4.18) can be recast into a momentum continuity statement

$$\frac{\partial}{\partial t}(\rho\dot{x}_i) - \frac{\partial}{\partial z_l}\left(t_{il}^E - \rho\dot{x}_i\dot{x}_j\right) = qE_i + (\mathbf{j}\times\mathbf{B})_i . \quad (5.2)$$

The momentum continuity equation for the electromagnetic field is obtained by forming a vector product of Eq. (4.2) with **B**, a vector product of Eq. (4.7) with **E**, a product of Eq. (4.1) with **E**, and a product of Eq. (4.8) with $\mathbf{B}/\mu_0$, and adding them to obtain

$$\frac{\partial}{\partial t}(\epsilon_0\mathbf{E}\times\mathbf{B})_i - \frac{\partial m_{il}}{\partial z_l} = -qE_i - (\mathbf{j}\times\mathbf{B})_i , \quad (5.3)$$

where the electromagnetic stress tensor is defined as

$$m_{il} = \epsilon_0 E_i E_l + \frac{1}{\mu_0}B_i B_l - \frac{1}{2}\left(\epsilon_0 E_k E_k + \frac{1}{\mu_0}B_k B_k\right)\delta_{il}. \quad (5.4)$$

The addition of Eqs. (5.2) and (5.3) gives the conservation law of momentum in the spatial frame

$$\frac{\partial}{\partial t}(\rho\dot{\mathbf{x}} + \epsilon_0\mathbf{E}\times\mathbf{B})_i - \frac{\partial}{\partial z_l}\left(t_{il}^E + m_{il} - \rho\dot{x}_i\dot{x}_l\right) = 0. \quad (5.5)$$

Note that neither the internal motion nor spin equations contribute to this conservation law as expected since neither can carry momentum.

C. Angular momentum conservation

We form an angular momentum continuity equation of each field since each is capable of possessing angular momentum. First, we form a vector product of **x** with Eq. (5.2), the modified form of the dynamic equation for the center-of-mass field. It becomes

$$\frac{\partial}{\partial t}\left[(\rho\mathbf{x}\times\dot{\mathbf{x}})_i\right] + \frac{\partial}{\partial z_l}\left[\rho(\mathbf{x}\times\dot{\mathbf{x}})_i\,\dot{x}_l - \epsilon_{ijk}x_j t_{kl}^E\right]$$
$$= -\epsilon_{ilk}t_{kl}^E + q(\mathbf{x}\times\mathbf{E})_i + [\mathbf{x}\times(\mathbf{j}\times\mathbf{B})]_i . \quad (5.6)$$

Second, we form a vector product of **x** with Eq. (5.3), the momentum continuity equation of the electromagnetic field, to obtain after some rearrangement

$$\frac{\partial}{\partial t}\left[\epsilon_0\mathbf{x}\times(\mathbf{E}\times\mathbf{B})\right]_i - \frac{\partial}{\partial z_l}(\epsilon_{ijk}x_j m_{kl})$$
$$= -q(\mathbf{x}\times\mathbf{E})_i - [\mathbf{x}\times(\mathbf{j}\times\mathbf{B})]_i . \quad (5.7)$$

Third, we form the vector product of $J^{-1}\mathbf{y}^{T\nu}$ with Eq. (4.10) and then sum it over ν for all the internal coordinates. We obtain

$$J^{-1}\frac{d}{dt}(Jl_i^i) = \frac{\partial l_i^i}{\partial t} + \frac{\partial}{\partial z_l}(l_i^i\dot{x}_l) = \epsilon_{ilk}P_l\mathcal{E}_k + 2\epsilon_{ilk}Q_{lm}\mathcal{E}_{k,m} + \epsilon_{ilk}J^{-1}\sum_{\mu\nu}q^{\mu\nu}\epsilon_{kmn}y_l^{T\nu}\dot{y}_m^{T\mu}B_n$$
$$-\epsilon_{ilk}\epsilon_{kmn}2Q_{lp}\dot{x}_{m,p}B_n - \epsilon_{ilk}J^{-1}\sum_\nu y_l^{T\nu}\frac{\partial\rho^0\Sigma}{\partial y_k^{T\nu}}$$
$$-\epsilon_{ilk}J^{-1}\sum_\nu y_{l,A}^{T\nu}\frac{\partial\rho^0\Sigma}{\partial y_{k,A}^{T\nu}} + \left(\epsilon_{ilk}J^{-1}\sum_\nu\frac{\partial\rho^0\Sigma}{\partial y_{k,A}^{T\nu}}y_l^{T\nu}x_{m,A}\right)_{,m} , \quad (5.8)$$

where l^i is the angular momentum density of internal motions in the spatial frame defined as

$$l^i = \sum_\nu \rho^\nu\mathbf{y}^{T\nu}\times\dot{\mathbf{y}}^{T\nu}, \quad (5.9)$$

with $\rho^\nu \equiv J^{-1}m^\nu$.

Fourth, we multiply Eq. (4.13) by J^{-1} and then sum it over α,

$$J^{-1}\frac{d}{dt}(Jl_i^s) = \frac{\partial l_i^s}{\partial t} + \frac{\partial}{\partial z_l}(l_i^s\dot{x}_l)$$
$$= \epsilon_{ilk}M_l^s B_k + \left(J^{-1}\epsilon_{ilk}\sum_\alpha\frac{\partial\rho^0\Sigma}{\partial s_{k,A}^{T\alpha}}s_l^{T\alpha}x_{m,A}\right)_{,m} - J^{-1}\sum_\alpha\epsilon_{ilk}s_l^{T\alpha}\frac{\partial\rho^0\Sigma}{\partial s_k^{T\alpha}} - J^{-1}\sum_\alpha\epsilon_{ilk}s_{l,A}^{T\alpha}\frac{\partial\rho^0\Sigma}{\partial s_{k,A}^{T\alpha}}, \quad (5.10)$$

where l^s is the spin angular momentum density in the spatial frame defined as

$$l^s = J^{-1}\sum_\alpha \mathbf{s}^{T\alpha}. \quad (5.11)$$

Finally we add the angular momentum continuity equations (5.6), (5.7), (5.8), and (5.10), together. After considerable manipulation and cancellation of terms this leads to the angular momentum conservation law in the spatial frame,

$$\frac{\partial}{\partial t}(\mathbf{x}\times\mathbf{g} + l^i + l^s)_i + \frac{\partial}{\partial z_l}\left[-\epsilon_{ijk}x_j t_{kl}^T + (l_i^i + l_i^s)\dot{x}_l \right.$$
$$\left. -\epsilon_{ijk}\frac{1}{J}\left(\sum_\nu\frac{\partial\rho^0\Sigma}{\partial y_{k,A}^{T\nu}}y_j^{T\nu} + \sum_\alpha\frac{\partial\rho^0\Sigma}{\partial s_{k,A}^{T\alpha}}s_j^{T\alpha}\right)x_{l,A}\right] = 0 , \quad (5.12)$$

where $\mathbf{g} \equiv \rho\dot{\mathbf{x}} + \epsilon_0\mathbf{E}\times\mathbf{B}$ is the total momentum density of the system and $t_{kl}^T \equiv t_{kl}^E + m_{kl} - \rho\dot{x}_k\dot{x}_l$ is the total stress tensor.[2] We note that all the dynamical equations contribute to the density and flow of angular momentum.

It is worth noting that a rearrangement of an intermediate step in the derivation of Eq. (5.12) yields an important statement concerning the asymmetry of the elastic stress tensor (4.19),

$$\frac{1}{J}\frac{d}{dt}\left[J(l_j^i + l_j^s)\right] = \epsilon_{jkl}\left[t_{lk}^E + (Q_{km}\mathcal{E}_l)_{,m}\right]. \quad (5.13)$$

This statement, without the effects of intrinsic spin and those from the magnetization and quadrupolarization arising from charge, has been discussed before.[2,27] Briefly it states that the elastic stress tensor and the quadrupolarization term in Eq. (5.13) can be asymmetric without violating angular momentum conservation. Instead, the torque created by the asymmetry causes a change in the internal angular momentum density of the internal motions and, as seen here for the first time, the intrinsic spins also. The new spin density term $\boldsymbol{l}^s$ can easily dominate the internal motion term $\boldsymbol{l}^i$ in ferromagnetic and antiferromagnetic crystals. This may have important implications in the choice of crystals in the search for violations of the Cauchy symmetry of the stiffness tensor near second-order phase transitions driven by a soft mode, an effect previously predicted.[27]

D. Energy conservation

To obtain the energy conservation law we form the continuity equation of energy for each field and then add them together. First, we form the scalar product of $\dot{\mathbf{x}}$ with the equation of motion of the center-of-mass field in the spatial frame, Eq. (4.18), to obtain

$$\rho\dot{x}_i\ddot{x}_i = J^{-1}\frac{d}{dt}\left(J\rho\dot{\mathbf{x}}^2/2\right) = \frac{\partial}{\partial t}\left(\rho\dot{\mathbf{x}}^2/2\right) + \frac{\partial}{\partial z_j}\left(\rho\dot{\mathbf{x}}^2\dot{x}_j/2\right) = \left(t^E_{ij}\dot{x}_i\right)_{,j} - t^E_{ij}\dot{x}_{i,j} + \dot{x}_i\left[qE_i + (\mathbf{j}\times\mathbf{B})_i\right]. \tag{5.14}$$

Second, we form the scalar product of $J^{-1}\dot{\mathbf{y}}^{T\nu}$ with the dynamic equation for the internal motion field, Eq. (4.10), and then sum it over the index ν. We obtain

$$\begin{aligned}
J^{-1}\sum_\nu m^\nu \dot{y}^{T\nu}_i\ddot{y}^{T\nu}_i &= J^{-1}\frac{d}{dt}\left(J\sum_\nu \rho^\nu\frac{1}{2}\left(\dot{\mathbf{y}}^{T\nu}_i\right)^2\right)\\
&= \frac{\partial}{\partial t}\left(\sum_\nu \rho^\nu\frac{1}{2}\left(\dot{\mathbf{y}}^{T\nu}_i\right)^2\right) + \frac{\partial}{\partial z_j}\left[\sum_\nu \rho^\nu\frac{1}{2}\left(\dot{\mathbf{y}}^{T\nu}_i\right)^2\dot{x}_j\right]\\
&= J^{-1}\sum_\nu q^\nu\dot{y}^{T\nu}_i\mathcal{E}_i + J^{-1}\sum_\nu q^{\nu\mu}\dot{y}^{T\nu}_i y^{T\nu}_j\mathcal{E}_{i,j}\\
&\quad -J^{-1}\sum_{\mu\nu} q^{\mu\nu}\dot{y}^{T\nu}_i\epsilon_{ijk}y^{T\mu}_l\dot{x}_{j,l}B_k - J^{-1}\sum_\nu \dot{y}^{T\nu}_i\frac{\partial\rho^0\Sigma}{\partial y^{T\nu}_i}\\
&\quad -J^{-1}\sum_\nu \dot{y}^{T\nu}_{i,A}\frac{\partial\rho^0\Sigma}{\partial y^{T\nu}_{i,A}} + \left(J^{-1}\sum_\nu\frac{\partial\rho^0\Sigma}{\partial y^{T\nu}_{i,A}}\dot{y}^{T\nu}_i x_{j,A}\right)_{,j}.
\end{aligned}\tag{5.15}$$

Third, we form the continuity equation for the electromagnetic field energy. By forming the scalar products of $-\mathbf{E}$ with Eq. (4.2) and $\mathbf{B}/\mu_0$ with Eq. (4.7), and then summing them, we obtain the familiar continuity equation for the electromagnetic field energy as

$$\frac{\partial}{\partial t}\left(\frac{\epsilon_0\mathbf{E}^2}{2} + \frac{\mathbf{B}^2}{2\mu_0}\right) + \boldsymbol{\nabla}\cdot\left(\frac{\mathbf{E}\times\mathbf{B}}{\mu_0}\right) = -\mathbf{E}\cdot\mathbf{j}. \tag{5.16}$$

Next we form the scalar product of $-J^{-1}\mu^\alpha\mathbf{B}$ with the dynamic spin equation (4.13) and sum it over α,

$$\begin{aligned}
-\frac{B_i}{J}\frac{d}{dt}\left(JM^s_i\right) &= -B_i\frac{\partial M^s_i}{\partial t} - \frac{\partial}{\partial z_l}\left(B_iM^s_i\dot{x}_l\right) + M^s_i\dot{x}_lB_{i,l}\\
&= \frac{1}{J}\sum_\alpha \mu^\alpha B_i s^{T\alpha}_j\epsilon_{ijk}\left(\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_k} - \frac{d}{dX_A}\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{k,A}}\right)\\
&= -\frac{1}{J}\sum_\alpha\left[\dot{s}^{T\alpha}_k + \epsilon_{klm}s^{T\alpha}_l\left(\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_m} - \frac{d}{dX_A}\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{m,A}}\right)\right]\left(\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_k} - \frac{d}{dX_A}\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{k,A}}\right)\\
&= -\frac{1}{J}\sum_\alpha\left(\dot{s}^{T\alpha}_k\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_k} + \dot{s}^{T\alpha}_{k,A}\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{k,A}}\right) + \left(\frac{1}{J}\sum_\alpha\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{k,A}}\dot{s}^{T\alpha}_k x_{l,A}\right)_{,l}.
\end{aligned}\tag{5.17}$$

By adding Eqs. (5.14), (5.15), (5.16), and (5.17), and after considerable manipulation we obtain the energy conservation law expressed in the spatial frame to be

$$\begin{aligned}
&\frac{\partial}{\partial t}\left(\frac{\rho\dot{\mathbf{x}}^2}{2} + \sum_\nu\frac{\rho^\nu}{2}\left(\dot{\mathbf{y}}^{T\nu}_i\right)^2 + \rho\Sigma - \mathbf{M}^s\cdot\mathbf{B} + \frac{\epsilon_0\mathbf{E}^2}{2} + \frac{\mathbf{B}^2}{2\mu_0}\right)\\
&+\frac{\partial}{\partial z_j}\left[\left(\frac{\rho\dot{\mathbf{x}}^2}{2} + \sum_\nu\frac{\rho^\nu}{2}\left(\dot{\mathbf{y}}^{T\nu}_i\right)^2 + \rho\Sigma - \mathbf{M}^s\cdot\mathbf{B}\right)\dot{x}_j - \left(t^E_{ij} + M_iB_j - \mathbf{M}\cdot\mathbf{B}\delta_{ij}\right)\dot{x}_i\right.\\
&\left.+\left(\mathbf{E}\times(\mathbf{B}/\mu_0 - \mathbf{M})\right)_j + \left(\dot{x}_{i,k}Q_{kj} - \frac{\partial Q_{ji}}{\partial t} - \left(Q_{ji}\dot{x}_k\right)_{,k}\right)\mathcal{E}_i - \frac{1}{J}\left(\sum_\nu\frac{\partial\rho^0\Sigma}{y^{T\nu}_{i,A}}\dot{y}^{T\nu}_i + \sum_\alpha\frac{\partial\rho^0\Sigma}{\partial s^{T\alpha}_{i,A}}\dot{s}^{T\alpha}_i\right)x_{j,A}\right] = 0.
\end{aligned}\tag{5.18}$$

It is interesting that the spin magnetization and magnetization from the motion of bound charge contribute differently to the energy density and to the flow of energy in the conservation law (5.18). The spin magnetization enters the energy density as if it were a (negative) stored energy in spite of its origin as an interaction energy in Eq. (3.4). The magnetization due to the motion of charge, like all other non-spin-interaction energy terms, does not contribute to the energy density. Note that the magnetization from the motion of charge described in our theory is due to the long-wavelength motion of (mostly ionic) charged particles which are affected by both macroscopic electric and magnetic fields. However, the atomic orbital magnetization, which can be viewed as originating from electrons rotating very fast around an ion, is not accounted for by the internal motion fields since such motions are certainly not long wavelength. Nevertheless, the atomic orbital magnetization has almost identical properties as the spin magnetization, as known from quantum mechanics. Therefore, we can treat it as spin magnetization. We surmise that our formalism should accommodate the atomic orbital magnetization by altering the gyromagnetic ratio in μ^α.

E. Pseudomomentum conservation

The pseudomomentum conservation law has been found to have important implications[8] concerning the Minkowski-Abraham controversy about the nature of the momentum density of a light wave in matter and also concerning the interpretation of related experiments.[28,29] Pseudomomentum conservation results from the homogeneity of a material body quite analogously to momentum conservation resulting from the homgeneity of space. For the pseudomomentum conservation law to be exact the body must be infinite in extent. However, if the body is very large compared to the interaction region or wavelength of the probe, it is a good approximation and hence useful.

The pseudomomentum conservation law can be found by (a) forming a scalar product of Eq. (4.17) with $-x_{i,C}$ over i, (b) forming a scalar product of Eq. (4.10) with $-y_{i,C}^{T\nu}$ over i and summing the latter over ν from l to $N-1$, (c) forming a scalar product of Eq. (4.12) from the left with $-i\xi_{i,C}^{T\alpha}$ over i and summing over α, and (d) adding the three contributions. Since the electromagnetic field is not material based, its equations do not contribute. After considerable manipulation a material frame ($\mathbf{X}, t$ independent variables) conservation law results in the form

$$\begin{aligned}
&\frac{d}{dt}\left[-\rho^0\dot{x}_i x_{i,C} - \sum_\nu m^\nu \dot{y}_i^{T\nu} y_{i,C}^{T\nu} - \tfrac{i}{2}\sum_\alpha \xi_i^{T\alpha}\xi_{i,C}^{T\alpha} + x_{i,C}\epsilon_{ijk}p_j B_k + x_{i,C}\epsilon_{ijk}q_{jl}B_{k,l} + \tfrac{1}{2}\epsilon_{ijk}\sum_{\mu\nu} q^{\mu\nu} y_{i,C}^{T\mu} y_j^{T\nu} B_k\right] \\
&\quad -\frac{d}{dX_A}\left\{-\delta_{CA}\left[\rho^0\dot{x}^2/2 + \sum_\nu m^\nu (\dot{y}^{T\nu})^2/2 + \tfrac{i}{2}\sum_\alpha \xi_i^{T\alpha}\dot{\xi}_i^{T\alpha} - \rho^0\Sigma \right.\right. \\
&\quad \left.\left. +\mathbf{p}\cdot(\mathbf{E}+\dot{\mathbf{x}}\times\mathbf{B}) + \mathbf{m}\cdot\mathbf{B} + q_{kl}(E_{k,l}+\epsilon_{kji}\dot{x}_j B_{i,l})\right] - x_{i,C}\frac{\partial\rho^0\Sigma}{\partial x_{i,A}} - \sum_\alpha s_{j,C}^{T\alpha}\frac{\partial\rho^0\Sigma}{\partial s_{j,A}^{T\alpha}}\right\} = 0 . \qquad (5.19)
\end{aligned}$$

It is interesting and somewhat surprising that the Grassmann variables appear in this equation rather than only spins.

The full implications of these Grassmann variable terms are not presently understood. Evaluating Eq. (5.19) for a linear light wave shows that the Grassmann term in the pseudostress tensor just cancels the spin part of $\mathbf{m}\cdot\mathbf{B}$. However, the Grassmann term in the pseudomomentum density becomes $\Sigma_\alpha \mu^\alpha \mathbf{S}^\alpha \cdot \mathbf{B} k_C/\omega$ where $\mathbf{k}$ and ω are the wave vector and frequency of the light wave. Evaluating the Grassmann terms of Eq. (5.19) for a spin wave appears more complicated with both such terms vanishing under high-symmetry conditions but not under lower-symmetry conditions. Further work on understanding these terms is necessary.

VI. DISCUSSION AND CONCLUSION

We believe the development of this paper presents a complete, fundamental, and natural solution to the problem of classical (i.e., long-wavelength or continuum) magnetism from intrinsic spin in dielectric crystals. It is complete because it includes in the formulation interactions of the spin magnetization with all the other excitations (acoustic, optic, and electromagnetic) of the medium at any order of nonlinearity constrained in form only by the conservation laws. It should be remarked that an optic mode, as used here, can include electronic or excitonic modes provided additional stored energy terms accounting for the wave vector dispersion effects of the small mass of such excitations are included. The development is fundamental because it introduces an internal property to the individual particles that gives rise to the intrinsic spin of the particle. Thus new degrees of freedom are introduced at the particle level and do not require imposition of a phenomenological constraint in contrast to previous introductions[13–18] of spin magnetization at the continuum level. Somewhat paradoxically we regard the development as natural by its introduction of an anticommuting Grassmann variable because the work of Berezin and Marinov[23] and Casalbuoni[24] showed that such a variable is the classical counterpart of a fermionic excitation. This also means that the reverse procedure of quantizing the classical theory containing Grassmann G_3 variables procedes directly by the Dirac method to the quantum theory of spin $\frac{1}{2}$.

The introduction of anticommuting quantities into classical physics could be considered either a contradiction in terms or another demonstration that intrinsic spin is entirely "nonclassical." However, since spin magnetization produces many continuum or classical phenomena and fermionic excitations require classical anticommuting variables, we believe that the term "classical physics" has been wrongly limited to commuting algebra and needs now an updated and expanded definition to include anticommuting algebra when appropriate. After all, the term "classical physics" has always been defined in retrospect.

It is an interesting result of this approach that, though the Lagrangian must be formulated in terms of the anticommuting Grassmann variables in order to account for spin magnetization, at the equations of motion level the spin, given as a vector product of Grassmann variables, can be introduced everywhere and a fully commuting theory is then obtained. An exception to this statement appears to be the pseudomomentum conservation law which contains terms in Grassmann variables unexpressible in terms of just spin. These new and interesting terms need further study and interpretation.

One caveat concerning our approach should be repeated. We have used Grassmann G_3 algebra in which the basic Grassmann variable is a real, three-component vector. Its quantization shows that it represents spin $\frac{1}{2}$. Many ferromagnetic and antiferromagnetic ions have "intrinsic spin" (total angular momentum quantum number) of higher values. We surmise that this can be handled by a change of the gyromagnetic ratio parameter in this theory. Exploration of which Grassmann algebra presumably corresponds to spin-$\frac{3}{2}$, -$\frac{5}{2}$, etc., is important to check this surmise but has not yet been carried out.

As discussed in the Introduction, this Lagrangian approach, to which the present spin work is a significant addition, has had a wide range of successes in finding significant errors in supposedly well-understood phenomena,[3-7] in helping resolve long standing controversies,[8,9] and in producing the first characterization of new nonlinear effects.[10-12] Thus, it is reasonable to extrapolate from that experience to hopes of comparable usefulness of the Lagrangian approach in magnetic phenomena. The field of magneto-optic interactions is the most natural place for initial applications of this theory because it has included all optic modes and their interactions that play a key role in such phenomena. Previous continuum formulations did not include optic modes and so are inappropriate for such studies. Quantum mechanical formulations, on the other hand, typically are constructed for a particular interaction of interest and thus are less likely to reveal unexpected effects that a general formulation as presented here may evidence. Also, they always introduce the deformation interaction phenomenologically.

Already at the conservation law level, the present work has revealed an interesting physical phenomenon: Magnetizations from the two types of sources, motion of charge and intrinsic spin, enter the energy density and the energy flow differently. This occurs even though the two forms of magnetization enter both the interaction Lagrangian and the Maxwell equations in the same manner. Thus this result arises from our inclusion of the equations of motion of each degree of freedom of the material medium in the system. This, of course, results from the Lagrangian representing a closed system of matter, electromagnetic field, and their interaction. This appears to be the first derivation of this difference in energetics between spin magnetization and magnetization from moving charge based on classical physics concepts. Comparable results have been obtained by de Groot and Suttorp[30,31] using semirelativistic quantum statistics. We are in agreement on the nonrelativistic terms.

Another new result found at the conservation law level is Eq. (5.13) which shows the important and apparently dominant role that the angular momentum density of intrinsic spin plays in balancing the torques created by antisymmetric stresses. At the linear level these antisymmetric stresses cause the loss of the Cauchy symmetry[27] of the stiffness tensor in dynamic interactions and thus to an elastic coupling to infinitesimal rotations. It has been proposed[27] that a Brillouin scattering study of a crystal near its second-order phase transition that is driven by a zone-center soft optic mode could verify the prediction. The significant involvement of intrinsic spin in Eq. (5.13) suggests that a qualifying ferromagnetic or antiferromagnetic crystal may be the optimum choice for study.

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