Volume Elements of Monotone Metrics on the $n \times n$ Density Matrices as Densities-of-States for Thermodynamic Purposes. II

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(April 1, 2022)

We derive explicit expressions for the volume elements of both the minimal and maximal monotone metrics over the $(n^2 - 1)$-dimensional convex set of $n \times n$ density matrices for the cases $n = 3$ and 4. We make further progress for the specific $n = 3$ maximal-monotone case, by taking the limit of a certain ratio of integration results, obtained using an orthogonal set of eight coordinates. By doing so, we find remarkably simple marginal probability distributions based on the corresponding volume element, which we then use for thermodynamic purposes. We, thus, find a spin-1 analogue of the Langevin function. In the fully general $n = 4$ situation, however, we are impeded in making similar progress by the inability to diagonalize a $3 \times 3$ Hermitian matrix and thereby obtain an orthogonal set of coordinates to use in the requisite integrations.

PACS Numbers 05.30.Ch, 03.65.-w, 02.50.-r

Keywords: quantum statistical thermodynamics, spin-1 systems, Langevin function, Brillouin function, monotone metrics, harmonic mean, prior probability, entangled spin-1/2 systems

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IV. CONCLUDING REMARKS

1. INTRODUCTION

In this communication, we report a number of results pertaining to a certain quantum-theoretic model of the thermodynamic properties of a system comprised of spin-1 particles, in particular, a small number of them. Our approach can be contrasted with the standard (Jaynesian [1–3]) one, which, “in some respects can be viewed as semiclassical [and] can presumably be justified when the number of the constituent particles is large — in which case the random phases can be averaged over . . . However, in the case of a small system . . . there seems to be no a priori
reason for adopting the conventional mixed state approach” [4]. Vigorous criticisms of the “orthodox information-theoretic foundations of quantum statistics” have been expressed by Band and Park in an extended series of papers [5]. Park [6] himself later wrote that “the details of quantum thermodynamics are presently unknown” and “perhaps there is more to the concept of thermodynamic equilibrium than can be captured in the canonical density operator itself.” Additionally, Lavenda [7] argued (as detailed in sec. II D) that there are deficiencies — from a probabilistic viewpoint — with the “Brillouin function” (used in the study of ferromagnetism), which is yielded by the standard methodology [4]. Contrastingly, Lavenda asserts that the “Langevin function” is free from such defects. (Aharoni [8, pp. 83,97,98], citing Yatsuya et al. [9], in support, reaches similar conclusions under an assumption of complete spatial isotropy and arbitrariness of the direction of the applied field.) We have previously found [10, App. I] (cf. [4]) that for the spin-\( \frac{1}{2} \) systems, the form of analysis we will pursue here for the spin-1 systems, does, in fact, yield the Langevin function. Thus, our methodology appears to be not subject to the criticism of Lavenda. One of the principal results below will be a spin-1 version (Fig. 4) of the (spin-\( \frac{1}{2} \)) Langevin function. One possible application of these results is to quantum chromodynamics, where one can regard the antiscreening of the Yang-Mills vacuum as paramagnetism for the gluons, which are charged particles of spin 1 [11,12].

In the context of entangled quantum systems — to which we seek to apply our analytical approach in sec. II A — it has been argued [13] that “there are situations where the Jaynes principle fails”. “The difficulties in understanding of the Jaynes inference scheme are due to the fact that the latter is just a principle and it was not derived within the quantum formalism” [14]. Friedman and Shimony [16], in a classical rather than quantum context, claim to have “exhibited an anomaly in Jaynes’ maximum entropy prescription,” cf. [17–19].

II. SPIN-1 SYSTEMS

A. Background

Bloore [15] had studied the geometrical structure of the eight-dimensional convex set of spin-1 density matrices, which he denoted,

\[
\rho = \begin{pmatrix}
    a & \bar{h} & g \\
    \bar{h} & b & \bar{f} \\
    g & \bar{f} & c
\end{pmatrix}, \quad a, b, c \in \mathbb{R}, \quad f, g, h \in \mathbb{C}
\]  

(we incorporate the notation of [15] into ours). In the (MATHEMATICA [20]) computations upon which we rely, a sequence of transformations — suggested by this work of Bloore — is implemented, leading to the full separation of the transformed variables [21]. In particular, we make use of a four-dimensional version \((r, \theta_1, \theta_2, \theta_3)\) of spheroidal coordinates [22].

In recent years, there have been several studies [23–27] concerning the assignment of certain natural Riemannian metrics to sets of density matrices, such as (1). These various metrics can all be considered to be particular forms of monotone metrics [28,29]. (Contrastingly, there is only classically, as shown by Chentsov [31], a single monotone metric — the one associated with the Fisher information. Morozova and Chentsov [32] sought to extend this work to the quantum domain, while Petz and Sudar [28] further developed the line of analysis.) Of particular interest are the maximal monotone metric (of the left logarithmic derivative) — for which the reciprocal \((\frac{\mu+\nu}{2})\) of the “Morozova-Chentsov function” [33] is the arithmetic mean — and the minimal (Bures) monotone metric (of the symmetric logarithmic derivative) — for which the reciprocal \((\frac{2\mu\nu}{\mu+\nu})\) of the Morozova-Chentsov function is the harmonic mean. Also of considerable interest is the Kubo-Mori metric [27] — the reciprocal \((\frac{\log \mu - \log \nu}{2})\) of the Morozova-Chentsov function of which is the logarithmic mean – but we do not study it here, if for no other reason than that it appears to be computationally intractable for our purposes. In any case, we should note that in [25], relying upon a variant of a statistical test devised by Clarke [34], the Kubo-Mori metric was found to give rise to a less noninformative prior distribution than the maximal monotone metric, which itself was shown to be most noninformative (but only if — due to nonnormalizability — the pure and some collection of nearly pure states, were eliminated from consideration). We will make use of the Morozova-Chentsov functions in deriving (by adopting certain work of Dittmann [35]) our formulas for the volume elements of the minimal and monotone metrics.
**B. Principal Results**

The first set of questions which we wish to address is whether one can normalize the volume elements of these two (minimal and maximal) metrics over the eight-dimensional convex set of spin-1 density matrices, so as to obtain (prior) probability distributions. In the maximal case (as also in its spin-1/2 counterpart [10, eqs. (43)-(46)]), the answer is strictly no, since we encounter divergent integrals. Nevertheless — similarly to the analysis in the spin-1/2 case — we are able to normalize the volume element of the maximal monotone metric over a subset of the entire convex set, omitting the pure and some collection of nearly pure states. Then, by taking the limit (in which the subset approaches the complete set) of a certain ratio, obtain a lower-dimensional marginal probability distribution.

In this manner, we have been able to assign the probability distribution (cf. (19)),

\[
\frac{15(1-a)\sqrt{a}}{4\pi \sqrt{b} \sqrt{c}},
\]

(2)

to the two-dimensional simplex spanned by the diagonal entries of (1). Of course, by the trace condition on density matrices, we have that \(a + b + c = 1\). The asymmetry under exchange that is evident between \(a\) and either \(b\) or \(c\) — but lacking between \(b\) and \(c\) themselves — in (2), is attributable to the specific sequence of transformations, suggested by the work of Bloore [15], employed below, following his notational and analytical scheme. Consequently, it is quite natural to interpret the variable \(a\) (despite the particular ordering of rows and columns used by Bloore in (1), whose notation we have adopted from the outset of our analysis) with the middle level of the three-level system (the one inaccessible to a spin-1 photon, due to its masslessness), and \(b\) and \(c\) with the other two (accessible to a two-level system). With another, but equally valid sequence of transformations, we could have interchanged the roles of these variables in (2). (Let us also note that without the factor, \(1 - a\), (2) would be proportional to a member of the Dirichlet family of distributions [36].)

The univariate marginal distributions of (2) are (Fig. 1) a member of the family of beta distributions,

\[
\frac{15(1-a)\sqrt{a}}{4},
\]

(3)

having a peak at \(a = \frac{1}{3}\), at which the probability density equals \(\frac{5}{\sqrt{3}} \approx 1.44338\) and (Fig. 2),

\[
\frac{15(1-b)(1+3b)}{32\sqrt{b}},
\]

(4)

(and similarly for the diagonal entry \(c\)). The expected values for these distributions are \(\langle a \rangle = \frac{2}{7}\), and \(\langle b \rangle = \langle c \rangle = \frac{2}{7}\), so \(\langle a \rangle + \langle b \rangle + \langle c \rangle = 1\). Also, \(\langle ab \rangle = \frac{2}{21}\), \(\langle a^2 \rangle = \frac{5}{21}\), \(\langle b^2 \rangle = \langle c^2 \rangle = \frac{1}{7}\).
C. Thermodynamic Analyses Based on Two Diagonal Hamiltonians

1. The first diagonal Case ($\lambda_8$)

Let us now consider the observable, one of a standard set (but, due to the particular sequence of transformations we employ, suggested by the work of Bloore [15], we take the liberty of harmlessly permuting the first and third rows and columns of the usual form of presentation) of eight Hermitian generators of $SU(3)$ [37,38],

$$\lambda_8 = \frac{1}{\sqrt{3}}\begin{pmatrix} -2 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix},$$

which might possibly function as the Hamiltonian of the spin-1 system. The expected value of (5) with respect to (1) is $\langle \lambda_8 \rangle = \text{Tr}(\rho \lambda_8) = \frac{1-3a}{\sqrt{3}}$. Multiplying the univariate marginal probability distribution (3) by the Boltzmann factor $\exp(-\beta \langle \lambda_8 \rangle)$ and integrating over $a$ from 0 to 1, we obtain the (“weak equilibrium” [5]) partition function (cf. [39]),

$$Q(\beta) = \frac{1}{16\beta^{3/2}}(5e^{-\beta} - 3\sqrt{\beta} - 3^{1/2}(2\beta + \sqrt{3})\text{erfi}(3^{1/2} \sqrt{\beta})),$$

where erfi represents the imaginary error function $\text{erfi}(z)$. (The error function erf($z$) is the integral of the Gaussian distribution, that is, $\frac{2}{\sqrt{\pi}} \int_0^z e^{-t^2} \, dt$.) In “strong equilibrium,” zero prior probability is assigned to those density matrices which do not commute with the Hamiltonian, while in “weak equilibrium,” this requirement is not imposed [3].

In the conventional manner of thermodynamics, we compute the expected value $(\langle\langle \lambda_8 \rangle \rangle)$ of $\langle \lambda_8 \rangle$ as $-\frac{2 \log Q(\beta)}{\beta}$. The result is plotted in Fig. 3.
The expected value for $\beta = 0$, corresponding to infinite temperature, is $\langle \lambda_8 \rangle = -\frac{1}{\sqrt{3}} \approx -0.5774$. Two physical conditions must be met in order for negative temperatures to arise: the subsystem possessing the negative temperature must be well insulated thermally from the other modes of energy storage of the complete system, and the subsystem energy levels must be bounded from above and below. Thus a normally populated state with probabilities proportional to $e^{-\beta E}$, can be inverted by reversal of the order of the energy levels while populations remain intact because there is no convenient energy sink available. Examination of the entropy gives further insight to the idea of negative temperatures. When the energy levels are bounded from above as well as below, however, zero entropy can occur for both minimum and maximum energy. No particular difficulties arise in the logical structure of statistical mechanics or of thermodynamics as a consequence of these negative-temperature systems.

Besides (5), the only diagonal member of the standard set of eight Hermitian generators of $SU(3)$ is

$$\lambda_3 = J_3^{(1)} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix},$$

(7)

where $J_3^{(1)}$ is used to denote a particular angular momentum matrix [46, p. 38]. This can be viewed as the spin-1 analogue of the Pauli matrix,

$$\sigma_3 = 2J_3^{(\frac{3}{2})} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}.$$

(8)

Use of this observable ($\sigma_3$) in conjunction with the volume element (inversely proportional to the $\frac{3}{2}$-power of the determinant of the $2 \times 2$ density matrix) of the maximal monotone metric for spin-$\frac{3}{2}$ systems, has led to the partition function,

$$Q(\beta) = \frac{\sinh(\beta)}{\beta},$$

(9)

yielding as the expected value, the negative of the Langevin function [47],

$$-\frac{\partial \log Q(\beta)}{\partial \beta} = \frac{1}{\beta} - \coth \beta.$$

(10)

2. The second diagonal case ($\lambda_3$)

Now, we have additionally that $\langle \lambda_3 \rangle = \text{Tr}(\rho \lambda_3) = b - c$. Then, using a combination of symbolic and numerical integration, we have conducted a thermodynamic analysis for $\lambda_3$, analogous to that above (5) and Fig. 3 for $\lambda_8$. In
Fig. 4, we display the expected value \( \langle \langle \lambda_3 \rangle \rangle \) of \( \langle \lambda_3 \rangle \), obtained numerically, along with that predicted by the negative of the Langevin function (10). The two curves are rather similar in nature, with the negative of the Langevin function assuming, in general, greater absolute expected values for a given \( \beta \).

![Graph showing expected values vs. inverse temperature parameter \( \beta \)](image)

**FIG. 4.** Expected value of \( \langle \lambda_3 \rangle \) as a function of the inverse temperature parameter \( \beta \), along with the expected value predicted by the (steeper-at-the-origin) negative of the Langevin function (10).

### D. Comparative properties of the Brillouin and Langevin functions

#### 1. Critique of Lavenda

The usual use of the Langevin function is to describe the thermodynamic behavior of noninteracting particles with nonquantized spin. In this same paradigm, the Brillouin function, that is \( \tanh \beta \), is employed for spin-\( \frac{1}{2} \) particles [47]. (Its spin-1 counterpart is \( \frac{2\sinh \beta}{1+2\cosh \beta} \).) However, Lavenda [7, pp. 193] has argued that, in contrast to the Langevin function, the Brillouin function lacks a proper probabilistic foundation, since its generating function cannot be derived as the Laplace transform of a prior probability density”. He also writes [7, p. 198]: “Even in this simple case of the Langevin function … we have witnessed a transition from a statistics dictated by the central-limit theorem, at weak-fields, to one governed by extreme-value distributions, at strong-fields. Such richness is not possessed by the Brillouin function, for although it is almost identical to the Langevin function in the weak-field limit, the Brillouin function becomes independent of the field in the strong-field limit. In the latter limit, it would imply complete saturation which does not lead to any probability distribution. This is yet another inadequacy of modeling ferromagnetism by a Brillouin function, in the mean field approximation.” Additionally, Lavenda asserts [7, p. 20] that the Langevin function “has empirically been targeted as providing a good description of hysteresis curves in ferromagnetic materials when the field due to interdomain coupling is added to the actual internal field to produce an effective field. This effective field is analogous to the Weiss mean field experienced by individual magnetic moments within a single domain. For a sufficiently large interdomain coupling parameter, an elementary form of hysteresis loop has been observed in the modified Langevin function. Moreover, since the generating function must be expressed as a Laplace transform of a prior distribution, in order to make physical as well as statistical sense, this rules out certain other candidates like the Brillouin function. For particles of spin-\( \frac{1}{2} \), the generating function would be proportional to the hyperbolic cosine, and the hyperbolic cosine cannot be expressed as a Laplace transform of a prior distribution.”

#### 2. The Jiles-Atherton theory

In his claims that the Langevin function provides a good description of certain magnetic phenomena, Lavenda refers to the work of Jiles and Atherton [48,49]. “The Jiles-Atherton theory is based on considerations of the dependence of energy dissipation within a magnetic material resulting from changes in its magnetization. The algorithm based on the theory yields five computed model parameters, \( M_s, a, \alpha, k \) and \( c \), which represent the saturation magnetization, the effective domain density, the mean exchange coupling between the effective domains, the flexibility of domain
walls and energy-dissipative features in the microstructure, respectively . . . The model parameter \( a \) is derived from an analogy to the Langevin expression for the anhysteretic magnetization \( M_{an} \) as a function of both temperature \( T \) and field \( H \) for a paramagnet . . . However, in the Jiles-Atherton theory, the spin entity \( \langle m \rangle \) is not an atomic magnetic moment \( m = n \mu_B \), where \( \mu_B \) is the Bohr magneton, as in the original Langevin expression. Rather, it represents the moment from a mesoscopic collections of spins that we refer to as an ‘effective domain;’ each ‘effective domain’ possesses a collective magnetic moment \( \langle m \rangle \). These effective domain entities may or may not correspond to actual magnetic domains” [50].

3. Critique of Brody and Hughston

Brody and Hughston [4, 51] propose the use of the negative of the Langevin function, that is \( \text{(10)} \), for the internal energy of a (small) system of spin-\( \frac{1}{2} \) particles in thermal equilibrium. Brody (personal communication) has suggested that “after all, standard (Einstein’s) approach to quantum statistical mechanics does seem to work for bulk objects, so there seems to be some kind of ‘transition’ from micro to macro scales”. “We note that in the case of the quantum canonical ensemble the heat capacity for this [spin-\( \frac{1}{2} \)] system is nonvanishing at zero temperature. Since it is known in the case of many bulk substances that the heat capacity vanishes as zero temperature is approached, it would be interesting to enquire if a single electron possesses a different behaviour, as indicated by our results” [4].

4. Critique of Aharoni

In sec. 5.2, entitled “Superparamagnetism” (that is, the “phenomenon of the loss of ferromagnetism in small particles”) of his recent text [8, pp. 97, 98], Aharoni writes: “A single particle of such a small size cannot be made or handled. Experiments are therefore carried out on an ensemble of particles, which in most cases have a wide distribution of particle sizes. Such particles would give rise to a superposition of Langevin functions with different values of \( \mu = M_s V \) in the argument, and the measured curve could not possibly look like the Langevin function . . . With improved techniques for producing very small particles, their size distribution has become narrow enough for a pure Langevin function [citing [9]] to be observed . . . The calculation [of the Langevin function] can now be said to have been confirmed by direct experiment. Of course, a Langevin function (or any other similar function) can always be fitted to such data for a rather narrow temperature range [citing [41, 42]], but the remarkably narrow distribution of [ [9]] can be fitted to such a function over a wide temperature range. In this respect this experiment is still quite unique in the literature” (cf. [43]).

Earlier [8, p. 84], Aharoni asserts: “And there is no mistake in this algebra: there are only two differences between this calculation [of the Langevin function] and the study of a gas of paramagnetic atoms in section 2.1 [yielding the Brillouin function]. One is that the function \( \theta \) [the angle to the fixed magnetic field] is continuous here, while this variable had discrete values in section 2.1 and the other is that the magnetic moment \( \mu \) was that of a single atom there, while here it is the moment of a large number of atoms, coupled together. However, the second difference is only quantitative and not qualitative, and the first one should not make any difference, especially since the energy levels of a large spin number \( S \) are very close together, and look like a continuous variable. It is thus true that if there was no other energy term besides the isotropic Heisenberg Hamiltonian, it would have been impossible to measure any magnetism in zero applied field, and there would be no meaning to a Curie temperature, or critical exponents, or any of the other nice features mentioned in the previous chapters. Theorists who calculate these properties never pay attention to the fact that the possibility of measuring that which they calculate is only due to an extra energy term, which they always leave out. Of course, a magnetization as in [the Langevin function], which is zero in zero applied field, contradicts not only experiments . . . It is also in conflict with everyday experience . . . It is because real magnetic materials are not isotropic, and not all values of the angle \( \theta \) are equally probable.”

5. Relations to modified Bessel functions

It is also interesting to observe that the Brillouin and Langevin functions are both instances \( (D = 1 \text{ and } D = 3, \text{ respectively}) \) of the two-spin correlation function of the D-vector model (of isotropically-interacting D-dimensional classical spins) for a one-dimensional lattice [58, Fig. 2.3],
where $I(x)$ is a modified Bessel function of the first kind.

**E. Derivation of the Volume Element of the Maximal Monotone Metric for Spin-1 Systems**

We now discuss the manner in which our first reported result \([13]\) was derived. To begin with, we noted that Bloore \([13]\) had suggested the transformations (“to suppress the dependence on [the diagonal entries] $a, b$ and $c$ by scaling the [off-diagonal] variables $f, g, h$),

$$f = \sqrt{bc} F, \quad g = \sqrt{ac} G, \quad h = \sqrt{ab} H.$$ \[(12)\]

The positivity conditions on the density matrix $\rho$, then, took the form \([13, \text{eq. (15)}]\),

$$|F| \leq 1, \quad |G| \leq 1, \quad |H| \leq 1, \quad |F|^2 + |G|^2 + |H|^2 - 2\text{Re}(FGH) \leq 1.$$ \[(13)\]

Bloore indicated that if $F = F_R + iF_I$ were simply real (that is, $F_I = 0$), then, the last condition could be rewritten as

$$1 - F_R^2 \geq G_R^2 + G_I^2 + H_R^2 + H_I^2 - 2F_R(G_RH_R - G_IH_I).$$ \[(14)\]

Then, by the rotation of the $(G_R, H_R)$-pair of axes through an angle of $\frac{\pi}{4}$ and the $(G_I, H_I)$-pair of axes, similarly, the allowed values of $G_R, H_R, G_I, H_I$ could be seen to lie inside a four-dimensional spheroid, two of the principal axes of which had a length equal to $\sqrt{1 + F_R}$ and the other two, $\sqrt{1 - F_R}$ (so the spheroid is neither predominantly “oblate” nor “prolate” in character). Bloore’s argument easily extends — using two additional identical rotations, not now necessarily however, equal to $\frac{\pi}{4}$ — to the general case, in which $F$ is complex. (If we were to single out $G$, say, rather than $F$, we would obtain an analogue of \[(3)\], containing $b$, not $a$, as its distinguished variable among $a, b$ and $c$.)

We proceed onward by converting to polar coordinates, $F_I = s \cos \nu, F_R = s \sin \nu$. Then, we rotate two of the six pairs formed by the four axes obtained by the two $\frac{\pi}{4}$-rotations — each new pair being comprised of one member from each of the two pairs resulting from the $\frac{\pi}{4}$-rotations suggested by Bloore. The two new angles of rotation now both equal $\frac{1}{2} \cot^{-1}(\tan \nu)$. We have, after performing the four indicated rotations, transformed the resultant set of axes $(J_1, J_2, J_3, J_4)$, using a set of four “hyperspherical” coordinates $(r, \theta_1, \theta_2, \theta_3)$,

$$J_1 = r \sqrt{1 + s \cos \theta_1}, \quad J_2 = r \sqrt{1 + s \cos \theta_2 \sin \theta_1},$$

$$J_3 = r \sqrt{1 - s \cos \theta_3 \sin \theta_2 \sin \theta_1}, \quad J_4 = r \sqrt{1 - s \sin \theta_3 \sin \theta_2 \sin \theta_1}.$$ \[(15)\]

The Jacobian of the total transformation (the scaling transformations \[(12)\], the four rotations of pairs of axes, along with the introduction of polar and hyperspherical coordinates) is,

$$J(a, b, c, s, r, \theta_1, \theta_2) = a^2 b^2 c^3 r^3 s(1 - s^2) \sin \theta_1^2 \sin \theta_2.$$ \[(16)\]

In the new variables, the determinant of the $3 \times 3$ density matrix $\rho$ takes the simple form (being free of the four angular variables — $u, \theta_1, \theta_2, \theta_3$),

$$|\rho| = abc(1 - r^2)(1 - s^2).$$ \[(17)\]

To arrive at the volume element of the maximal monotone metric, which we seek to integrate over the eight-dimensional convex set of spin-1 density matrices, we adopted an observation of Dittmann \([13]\) regarding the eigenvalues of the sum $(L_{\rho} + R_{\rho})$ of the operators of left $(L_{\rho})$ and right multiplication $(R_{\rho})$ for the minimal monotone metric. He had noted that for the $n \times n$ density matrices, in general, these $n^2$ eigenvalues would be of the form $p_i + p_j$ ($i, j = 1, \ldots, n$), where the $p_i$ ($i = 1, \ldots, n$) are the eigenvalues of $\rho$ itself. Then, we observed \([12]\) (cf. \([13, \text{eq. (24)}]\)) — making use of the fact that the determinant of a matrix is equal to the product of eigenvalues of the matrix — that the corresponding volume element would be proportional to the square root of the determinant of $(L_{\rho} + R_{\rho})^{-1}$ (or, equivalently, to the reciprocal of the square root of the determinant of $L_{\rho} + R_{\rho}$). We adopted this line of argument to the maximal monotone case by replacing the sums $p_i + p_j$ by (twice) the corresponding harmonic means.
Then, the volume element of the maximal monotone metric is proportional to \(|\rho|^{-\frac{5}{2}}\). Its use for thermodynamical purposes, led \([57]\) to a somewhat different ratio (cf. \([1, -I_2(\beta)/I_1(\beta)\), of modified Bessel functions, than the negative of the Langevin function \([10]\), which is expressible \([58]\) as \(-I_{3/2}(\beta)/I_{1/2}(\beta)\). (Brody and Hughston \([10]\) had, first, arrived at the former ratio, but then \([55]\) [personal communication], by using “phase space volume” rather than “state density” [i.e., weighted volume]”, concluded that the latter ratio was more appropriate. However, for higher dimensional cases — spin-1, the case under investigation here, being the simplest such — Brody and Hughston \([10]\) stated that “the energy surfaces are not fully ergodic with respect to the Schrödinger equation, and thus we cannot expect to be able to deduce the microcanonical postulate directly from the basic principles of quantum mechanics.”) So, it can be seen that as we pass from the spin-\(\frac{1}{2}\) case to the spin-1 situation, for both the minimal and maximal monotone metrics, we have to deal with an additional term (besides a certain power of \(|\rho|\)) of the form, \(w_2 - \rho\), to some power.

To find the bivariate marginal probability distribution \([10]\), we first performed the eightfold multiple integration (using the trace condition to set \(c = 1 - a - b\)),

\[
\int_0^1 \int_0^{1-a} \int_0^S \int_0^R \int_0^\pi \int_0^{2\pi} \int_0^{2\pi} |\rho|^{-\frac{5}{2}} (w_2 - |\rho|) J(a, b, c, s, r, \theta_1, \theta_2) d\nu d\theta_3 d\theta_2 d\theta_1 d\nu d\theta_1. \tag{18}
\]

Then, we (following established Bayesian principles for dealing with nonnormalizable prior distributions \([24, 25]\)) took the ratio of the outcome of the (initial sixfold) integration — that is, after all but the last two stages (over \(a\) and \(b\)) — to the result of the complete integration. In the double limit, \(R \to 1, S \to 1\), the ratio converges to the probability distribution \([11]\) over the two-dimensional simplex. (The result was invariant when the order in which the limits were taken was reversed.) This computational strategy was made necessary due to the fact that the integration diverged if we directly used the naive upper limits, \(r = 1\) and \(s = 1\), evident from \([11]\). A similar approach had been required (due to divergence also) in the spin-\(\frac{1}{2}\) case, based on the maximal monotone metric \([10]\, \text{eqs. (43)-(46)}\]. There, the corresponding volume element \((\propto |\rho|^{-\frac{5}{2}}\) was integrated over a three-dimensional ball of radius \(R\). As \(R \to 1\), this ball coincides with the “Bloch sphere” (unit ball) of spin-\(\frac{1}{2}\) systems. A bivariate marginal probability distribution was then obtainable by taking the limit \(R \to 1\) of a certain ratio — with the resultant univariate distribution simply being uniform in nature, and leading, when adopted as the density-of-states, to the Langevin model \([10]\). Since it has been argued elsewhere \([53, 54]\) that of all the possible monotone metrics, the maximal one is the most noninformative in character, it would seem plausible that the maximal metric might be singled out to provide density-of-states (structure) functions for thermodynamic analyses. (For additional results pertinent — in the context of “universal quantum coding” — to comparative properties of the maximal and monotone metrics, see \([22]\).)

We would also like to report a success in performing the eightfold multiple integration in \([18]\), after reordering the individual integrations so that the ones over \(r\) and \(s\) — rather than \(u\) and \(\theta_3\) — were performed first. Then, we were able to compute the limit of the ratio of the result after the first two integrations to the complete eightfold integration. This gives us a six-dimensional marginal probability distribution (free of \(r\) and \(s\), and independent of the particular values of \(\nu\) and \(\theta_3\)),

\[
\frac{15(1 - a)\sqrt{a} \sin \theta_1^4 \sin \theta_2^3}{8\pi^4 \sqrt{b} \sqrt{c}}. \tag{19}
\]

One of the two-dimensional marginal probability distributions of \((19)\) — the one over the simplex spanned by the three diagonal entries \((a, b, c)\) of the density matrix \([1]\) — is our earlier result \([2]\).

The focus of the study here has been on the application of symbolic integration to the modeling of the thermodynamics properties of spin-1 systems. With the application of numerical integration methods, however, we might hope to broaden our work to encompass non-diagonal Hamiltonians (such as \(\lambda_1, \lambda_2, \lambda_4, \lambda_5, \lambda_6, \lambda_7\)).
F. Thermodynamic Analysis Based on a Non-Diagonal Hamiltonian ($\lambda_1$)

Let us proceed with a “strong equilibrium” analysis [5] of the non-diagonal observable,

$$\lambda_1 = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \quad (20)$$

(It would appear that all our conclusions of a thermodynamic nature should be precisely the same for the other five non-diagonal observables, as well.) Rather than considering the full eight-dimensional convex set of $3 \times 3$ density matrices, however, we restrict our considerations to the two-dimensional convex set of $3 \times 3$ density matrices which commute with $\lambda_1$ — so, we are considering the case of “strong equilibrium” [5]. We, then, assign the volume element of the Bures (minimal monotone) metric to this set (using formula (10) in [23]), as a density-of-states. (Since the set is composed of commuting matrices, we are in a classical situation in which there is a unique monotone metric [28], so there is, in fact, no distinction between maximal and minimal ones, as earlier.) Our partition function then takes the form,

$$Q(\beta) = \int_0^1 \int_0^{1-g} \frac{e^{-\beta(1-2g-h)}}{2 \sqrt{g} \sqrt{h} \sqrt{1-g-h}} \, dh \, dg. \quad (21)$$

(We perform the inner integration symbolically, and the outer one, numerically.) In Fig. 5 (cf. Fig. 4), we plot the associated expected value, along with that given by the negative of the Langevin function [10].

![Graph](image)

**FIG. 5.** Expected value of $\langle \lambda_1 \rangle$ and the negative of the Langevin function [10].

Since these two curves quite remarkably almost coincide, in Fig. 6 we plot the result obtained by subtracting from the negative of the Langevin function [10], the expected value of $\lambda_1$. 

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III. SPIN-$\frac{3}{2}$ OR COUPLED SPIN-$\frac{1}{2}$ SYSTEMS

A. Required Transformations of the Fifteen Parameters

The possibility of extending our line of analysis to the $(n^2 - 1)$-dimensional convex sets of $n \times n$ density matrices, for $n > 3$, obviously, remains to be fully investigated. The $3 \times 3$ density matrix $\mathbb{1}$ can be considered to be embedded in the upper left corner of the $n \times n$ density matrix. Then, for this block, we can employ precisely the same set of transformations as in the analysis above. (Of course, there are $\frac{2(n-1)}{n}$ ways in which to actually position the $3 \times 3$ block. Depending upon which we choose, we would expect to break certain symmetries between the diagonal entries, as we witnessed with the probability distribution (3).) This leaves us with $n - 3$ new diagonal entries and $n^2 - 10$ new off-diagonal variables.

For the case $n = 4$ (which we had previously studied $[10]$ for various scenarios in which most of the fifteen parameters were $ab\ initio$ set to zero),

$$
\rho = \begin{pmatrix}
a & \bar{h} & \bar{g} & \bar{o} \\
\bar{h} & b & f & p \\
\bar{g} & f & c & \bar{q} \\
\bar{o} & \bar{p} & \bar{q} & d
\end{pmatrix}, \quad a, b, c, d \in \mathbb{R}, \quad f, g, h, o, p, q \in \mathbb{C}
$$

we have found that in the determinant, the cross-product between the real and complex parts of each new (scaled) off-diagonal entry (that is, $o, p, q$) is zero. Then, the extension of our earlier reasoning led us to form two triads, one element of each triad coming from one of these three pairs, and attempt to rotate them (in three-dimensional space), so as to eliminate the non-zero cross-product terms $[13-16]$. We have also investigated the possibility that by rotating the six variables in question in six-dimensional space, one could, in a single process, nullify all the non-zero cross-product terms. To accomplish this, it would be necessary to diagonalize a $6 \times 6$ symmetric matrix (or, equivalently, a $3 \times 3$ Hermitian matrix). The three pairs of equal eigenvalues of this matrix are given by the solution of the cubic equation,

$$
x^3 + \frac{1}{2}((6 - 2r^2 - r^2s - 2s^2 - r^2s(1 - 4\sin \theta_1^2 \sin \theta_2^2))x^2 + 3(1 - r^2)(1 - s^2)x + (1 - r^2)(1 - s^2)^2 = 0.
$$

(23)

There does not appear to be any very simple general form for the three roots of this equation. (In principle, we are able to diagonalize the $6 \times 6$ matrix, but the computations required to fully implement the associated transformations of the six variables, seem to be quite daunting.) However, if we set the factor $(1 - 4\sin \theta_1^2 \sin \theta_2^2)$ to zero (which can be accomplished by taking either $\theta_1$ or $\theta_2$ to be zero), then the three roots are of the form

$$
x = (-1 + r^2)(1 + s), \quad x = \frac{1}{2}(1 - s)(-2 - s + p), \quad x = -\frac{1}{2}(1 - s)(2 + s + p),
$$

(24)
where
\[ p = \sqrt{s^2 + 4r^2(1+s)}. \] (25)

If we set the factor to be -3 (by taking both \( \theta_1 \) and \( \theta_2 \) to be \( \frac{\pi}{2} \)), then the roots are
\[ x = (-1 + r^2)(1 - s), \quad x = \frac{1}{2}(1 + s)(-2 + s + p), \quad x = -\frac{1}{2}(1 + s)(2 - s + p), \] (26)
where now
\[ p = \sqrt{s^2 + 4r^2(1-s)}. \] (27)

The situation becomes simpler still if we set the parameter \( s \) to zero (so that the \( 2 \times 2 \) submatrix of \( (\mathbb{I}) \) in the upper left corner is diagonal in nature). Then, the three roots are \(-1 - r, -1 + r \) and \(-1 + r^2\). Similarly, for \( r = 0 \), the roots are \(-1 - s, -1 + s \) and \(-1 + s^2\). If we set \( s = 1 \) (corresponding to a pure state), two of the roots are zero and the third is \(-2(1 - r^2 + r^2 \sin \theta_1^2 \sin \theta_2^2)\). For \( r = 1 \), there are also two zero roots and one equalling \(-2 + s + s^2 - 2s \sin \theta_1^2 \sin \theta_2^2\).

Let us proceed to analyze in detail the case \( r = 0 \), for which the three eigenvalues, as just noted, are \(-1 - s, -1 + s \) and \(-1 + s^2\). Setting \( r \) to zero is equivalent (cf. (17)) to nullifying the entries \( g \) and \( h \) in (22), so the scenario is eleven-dimensional in nature. We have found the eigenvectors corresponding to these three eigenvalues, that is, for \((1 - s)\),
\[ (0, 0, \frac{\cos \nu}{\sqrt{2}}, -\sin \frac{\nu}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}}), \quad (0, 0, \frac{\sin \nu}{\sqrt{2}}, \frac{\cos \nu}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0), \] (28)
for \((-1 + s)\),
\[ (0, 0, -\frac{\cos \nu}{\sqrt{2}}, \frac{\sin \nu}{\sqrt{2}}, 0, \frac{1}{\sqrt{2}}), \quad (0, 0, -\frac{\sin \nu}{\sqrt{2}}, -\frac{\cos \nu}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0), \]
and for \((-1 + s^2)\),
\[ (0, 1, 0, 0, 0, 0), \quad (1, 0, 0, 0, 0, 0). \]

We transformed the three complex variables \( a, p \) and \( q \), on the basis of these eigenvectors, to a new set of six real variables \( (K_1 \ldots, K_6) \), between which there are no nonzero cross-product terms in the determinant of the transformed matrix. Then, we employed a six-dimensional version of spheroidal coordinates \((v, \xi_1, \xi_2, \xi_3, \xi_4, \xi_5)\) (cf. (13)),
\[ K_1 = v\sqrt{1 + s \cos \xi_1}, \quad K_2 = v\sqrt{1 + s \cos \xi_2 \sin \xi_1}, \]
\[ K_3 = v\sqrt{1 - s \cos \xi_3 \sin \xi_2 \sin \xi_1}, \quad K_4 = v\sqrt{1 - s \cos \xi_4 \sin \xi_3 \sin \xi_2 \sin \xi_1}, \]
\[ K_5 = v \cos \xi_5 \sin \xi_4 \sin \xi_3 \sin \xi_2 \sin \xi_1, \quad K_6 = v \sin \xi_5 \sin \xi_4 \sin \xi_3 \sin \xi_2 \sin \xi_1. \]

The determinant becomes, then, simply \( abcd(1 - s^2)(1 - v^2) \). The Jacobian corresponding to the sequence of transformations is (cf. (13))
\[ J(a, b, c, d, s, v, \xi_1, \xi_2, \xi_3, \xi_4) = ab^2 c^2 d^3 s(1 - s^2) v^5 \sin \xi_1^4 \sin \xi_2^3 \sin \xi_3^2 \sin \xi_4. \] (30)

**B. Volume Elements of the Maximal and Minimal Monotone Metrics for Spin-\(\frac{3}{2} \) Systems**

The integrand based on the maximal monotone metric is (employing once again our ansatz based on the work of Dittmann [35], in which we replace the arithmetic means of the eigenvalues by their harmonic means, in accordance with the corresponding Morozova-Chentsoff functions), the product of this Jacobian and the ratio of the term \( (w_2 w_3 - w_2^2 - |\rho|) \) to \( |\rho|^2 \), where \( w_2 \) is the sum of the six principal minors of order two and \( w_3 \) is the sum of the four principal minors of order three. We performed the integration over the eleven-dimensional convex set, the innermost integrations being over \( v \) from 0 to \( V \), and \( s \) from 0 to \( S \). (The integration would diverge if we were simply to set either \( S \) or \( V \)

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Then, we were able to integrate over the six angular variables \((u, \xi)'s\), but not fully over the three-dimensional simplex spanned by the diagonal entries \((a, b, c, d)\).

The problem, in this regard, seems attributable to the fact that the powers of \(a, b\) and \(c\) — but not \(d\) — in the Jacobian \((30)\) are all less than three. If all four powers were, in fact, equal to three, then dividing by \(|\rho|^{7/2}\), with \(|\rho| \propto abcd\), would yield a term of the form \((abcd)^{-7/2}\) — similar to the term \((abc)^{-1/2}\), encountered in the successful spin-1 analysis above — which would be integrable over the simplex. (We would, in fact, obtain such a desirable term if we could otherwise proceed with a full fifteen-dimensional analysis.) However, we have the resultant term \(a^{-7/2}b^{-5/2}c^{-5/2}d^{-1/2}\), which appears to be not integrable. Thus, we have not so far been able to extend the form of analysis taken here for the spin-1 systems to the convex set of spin-\(3/2\) or, equivalently, coupled spin-\(1/2\) systems (cf. [10]). (Following the lead of Bloore [13], one might also study the presumably simpler case of the nine-dimensional convex set of \(4 \times 4\) density matrices.)

C. Strong Equilibrium Thermodynamic Analysis of a Non-Diagonal Hamiltonian

Let us, in the manner of sec. II F, restrict our attention to those \(4 \times 4\) density matrices which commute with a certain non-diagonal Hamiltonian (so we remain within the framework of “strong equilibrium”), which we choose here to be

\[
\begin{pmatrix}
0 & 1 & 0 & 0 \\
1 & 0 & 1 & 0 \\
0 & 1 & 0 & 1 \\
0 & 0 & 1 & 0 \\
\end{pmatrix}.
\]

Then, after assigning the volume element of the Bures metric to the three-dimensional convex set of the mutually commuting density matrices, we have found the expected value of the observable \((31)\), as a function of the thermodynamic parameter \(\beta\), to be as indicated in Fig. 7.

![FIG. 7. Expected value of the non-diagonal observable (31)](image)

IV. CONCLUDING REMARKS

In this communication, we have sought to pursue a particular chain of reasoning (which, for spin-\(1/2\) systems, is known to yield the Langevin function, thus according fully with detailed arguments of Lavenda [7]) for spin-1 and spin-\(3/2\) systems. The fundamental problems encountered \(en route\) include the derivation of the volume elements of the (noninformative) maximal monotone metric (of the left logarithmic derivative) for these two scenarios. By applying principles used in Bayesian reasoning [59–61], we have succeeded through extensive analysis and computations in obtaining a six-dimensional marginal probability distribution \((19)\) from the (nonnormalizable) volume element for the
eight-dimensional convex set of spin-1 systems (sec. II). We have not been quite as successful, however, in addressing
the spin-$1/2$ scenario, in which the corresponding convex set is fifteen-dimensional in nature (sec. III).

We are not able to actually demonstrate, but can only conjecture, that our results have physical implications (cf.
[4]). The most questionable and debatable assumption underlying our analysis, perhaps, is that the volume element
of the maximal monotone metric can be employed as a density-of-states for thermodynamic purposes.

The initial impetus to pursue this line of research was provided by an extended series of forcefully-argued papers
of Band and Park [5]. In them, for various conceptual reasons, they expressed dissatisfaction with the conventional
(“Jaynesian”) approach to quantum statistical thermodynamics and sought to develop a conceptually superior alter-
native approach. We believe that our analysis is quite consistent with the spirit of their work, and serves to implement
it for certain low-dimensional scenarios. We have also been encouraged in our work by the detailed remarks of Lavenda
[7], regarding the propriety of using the Langevin function for spin-$1/2$ systems, since our methodology, in fact, yields
this function. Also, the analyses of Brody and Hughston [4,51], though differing from ours in several technical respects
— in particular, focusing on pure states — possess similar motivations.

It also appears natural that, amongst the continuum of monotone metrics [27,28], the maximal one should play the
distinguished information-theoretic role we have explored for it, since its noninformative nature has been independently
established [55,56]. The fact that, as a result of lengthy and demanding computations based on the volume element of
the maximal monotone metric, we were able to arrive at the quite simple results (2) and (19) also assists, we believe,
in validating the rather unconventional course pursued here.

ACKNOWLEDGMENTS

I would like to express appreciation to the Institute for Theoretical Physics for computational support in this
research and to Christian Krattenthaler for an illuminating discussion.

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