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Continuous families of isospectral Heisenberg spin systems and the limits of inference from measurements

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Abstract

We investigate classes of quantum Heisenberg spin systems which have different coupling constants but the same energy spectrum and hence the same thermodynamical properties. To this end we define various types of isospectrality and establish conditions for their occurrence. The triangle and the tetrahedron whose vertices are occupied by spin $\frac{1}{2}$ are investigated in some detail. The problem is also of practical interest since isospectrality presents an obstacle to the experimental determination of the coupling constants of small interacting spin systems such as magnetic molecules.

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1. Introduction

The measurement of the temperature-dependent magnetic susceptibility, $\chi(T)$, provides a standard essential diagnostic method for establishing the magnetic properties of a system. A careful comparison between measured data and the predictions for $\chi(T)$ as derived from a model Hamiltonian is routinely performed with the goal of establishing numerical values of model parameters, for example, the exchange constant(s) of the Heisenberg model of interacting isolated spins. The success of this technique is so firmly established that it is taken for granted that there is a one-to-one correspondence between a given form of $\chi(T)$ and the numerical values of the model parameters. Most certainly it is unnatural to contemplate that one might be able to continuously vary the parameters of a model Hamiltonian and yet generate a single, invariant form for $\chi(T)$ and similarly for other thermodynamic quantities. Yet surprisingly, there are a number of exceptional systems, where there is a continuous infinity-to-one correspondence between model Hamiltonians and measurable thermodynamic quantities. One of these exceptional cases was recently encountered [1, 2] in the course

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of attempting to determine the exchange constants of a simulating Heisenberg model from experimental susceptibility data for a specific synthetic magnetic compound [3].

We shall refer to systems of a continuous family having the same eigenvalue spectrum as being ‘isospectral’. This notion is chosen in analogy with the use of ‘isospectrality’ in other areas of physics, e.g. the occurrence of supersymmetric pairs of Hamiltonians (see, e.g., [4], section 7.1) or the problem of bounded domains with isospectral Laplacians (‘Can one hear the shape of a drum?’ [5]). The subject of isospectral spin systems is not completely novel but has been discussed in the literature only on a few occasions, e.g. [6, 7]. However, there is, to the best of our knowledge, no systematic account of this phenomenon, the first steps of which will be presented in this paper.

In addition to providing a general approach to isospectrality we analyse in depth two cases of distinct Heisenberg systems where continuous variation of the exchange constants gives rise to one and the same set of temperature-dependent thermodynamic quantities. The operational conclusion for an experimentalist is quite sobering, in that for these specific systems measured data alone cannot fix the exchange constants. Comparison between theory and experiment can only place a weak constraint on a continuous family of equally acceptable choices of parameters. Although we provide some helpful insights, it is very difficult to formulate the general set of conditions to be met so as to achieve such exceptional model systems. It is reasonable to expect that if one and the same temperature-dependent thermodynamic quantity is generated by a continuous family of Hamiltonians then necessarily all members of that family share the very same eigenvalue spectrum. This is indeed the case as is proven in section 3 for two particular thermodynamic functions.

In short, our goal in this paper is to provide a first systematic study of Heisenberg isospectral spin systems. The remarkable advances [8, 9] in synthesis magnetochemistry, of incorporating significant numbers of interacting paramagnetic centres within individual molecules, may provide the impetus for wider studies that will yield a more comprehensive set of conditions for the occurrence of isospectral spin systems.

The layout of this paper is as follows. In section 2 we introduce our notation and the basic concepts of ‘isospectrality’, ‘complete isospectrality’ and ‘covariant isospectrality’ for spin systems with Heisenberg Hamiltonians. Families of isospectral systems are algebraic varieties in the space \mathcal{J} of coupling constants. Covariant isospectrality is implemented by a unitary representation of some Lie group, which simplifies the calculations considerably. Unfortunately this is a rare case, as we will see. Complete isospectrality means that all eigenvalues of two systems with the same magnetic quantum number are in one-to-one correspondence and equal. We do not know whether this is a strictly stronger property than plain isospectrality, except for the case of a trivial counter-example. However, we need this apparently stronger concept to derive the conclusion that completely isospectral systems share the same magnetic susceptibility function. This is done in section 3 where the inverse problem is also settled as well as the analogous question for the specific heat function. The result in short is the following: plain isospectrality is equivalent to possessing the same specific heat function and is necessary for possessing the same magnetic susceptibility function. Complete isospectrality is sufficient for possessing the same magnetic susceptibility function. In section 4 we identify the isospectral invariants which are linear or quadratic in the coupling constants. This is crucial for section 5 where we show that the triangle with spin $s = \frac{1}{2}$ is both of completely and covariantly isospectral type but that for $s > \frac{1}{2}$ breaks down isospectrality. On the other hand, if the number N of spin sites exceeds 3, covariant isospectrality is no longer possible. This is proved in section 6 with the aid of MATHEMATICA 4.0 and some trace formulae which are explained in appendix A. The tetrahedron ($N = 4$) with $s = \frac{1}{2}$ nevertheless possesses completely isospectral families of dimension one and two, as shown in

section 7 and appendix B. In section 8 we provide some heuristic arguments in order to explain the findings of the previous sections. We expect that isospectrality only occurs if the number of all possible bonds exceeds the number of independent eigenvalues and show that this never happens except for $s = \frac{1}{2}$ and $N = 3, 4, 5$. A table summarizing our results and conjectures on the occurrence of isospectrality for different N and s and concluding remarks are provided in section 9.

2. Notation and definitions

We consider spin systems with N spin sites, spin quantum number s and isotropic Heisenberg coupling between all sites x and y with coupling constants J_{xy} . For the sake of compact notation we will write the $\binom{N}{2}$ coupling constants J_{xy} as the components of a vector $\mathbf{J} \in \mathcal{J} = \mathbb{R}^{\binom{N}{2}}$. Thus a specific point of \mathcal{J} uniquely specifies the strength of the interactions between all pairs of spins and will be sometimes called a ‘system’.

Let $S_x^{(i)}$ ($i = 1, 2, 3$), denote the three components of the spin observable S_x at site x and, as usual,

$$\mathbf{S} = \sum_x \mathbf{S}_x \quad S^{(i)} = \sum_x S_x^{(i)} \quad S^\pm = S^{(1)} \pm iS^{(2)} \tag{1}$$

denote the total spin vector and its various components. All linear operators occurring in this context will be identified with the corresponding $\text{dim} \times \text{dim}$ -matrices, $\text{dim} = (2s + 1)^N$ being the dimension of the total Hilbert space of the spin system, with respect to the fixed basis consisting of tensor products of eigenvectors of $S_x^{(3)}$. The Hamilton operator can then be written as

$$H_0 = \mathbf{J} \cdot \mathbf{H} = \sum_{x < y} J_{xy} H_{xy} \tag{2}$$

where

$$H_{xy} = \mathbf{S}_x \cdot \mathbf{S}_y = \sum_{i=1}^3 S_x^{(i)} S_y^{(i)}. \tag{3}$$

Here \mathbf{H} is an $\binom{N}{2}$ -dimensional vector, the components of which are $\text{dim} \times \text{dim}$ -dimensional matrices H_{xy} .

If the spin system is coupled to a constant external magnetic field \mathcal{H} , the total Hamilton operator will be

$$H(h) = H_0 - hS^{(3)} \tag{4}$$

where $h \equiv g\mu_B\mathcal{H}$ contains the common combination of the gyromagnetic ratio g and the Bohr magneton μ_B . As usual, the partition function, which yields all the standard thermodynamic functions, is defined by

$$\mathcal{Z}(\beta, h) \equiv \text{Tr}(\exp(-\beta H(h))). \tag{5}$$

In particular, one obtains from \mathcal{Z} the specific heat function

$$c(\beta) = \beta^2 \frac{\partial^2}{\partial \beta^2} \ln \mathcal{Z}(\beta, 0) \tag{6}$$

and the magnetic zero-field susceptibility function

$$\chi(\beta) = \frac{1}{\beta} \frac{\partial^2}{\partial h^2} \ln \mathcal{Z}(\beta, h)|_{h=0}. \tag{7}$$

Furthermore, we will need the traces of powers of H_0 , $t_n \equiv \text{Tr}(H_0^n)$, $n = 0, 1, 2, \dots$, \dim , and the set of traces

$$\mathcal{T}(H_0) = \{t_n | n = 0, 1, 2, \dots, \dim\}. \quad (8)$$

Two Hamilton operators with the same N and s , $H_0^{(1)} = \mathbf{J}^{(1)} \cdot \mathbf{H}$ and $H_0^{(2)} = \mathbf{J}^{(2)} \cdot \mathbf{H}$ are called *isospectral* if they have the same eigenvalues, counted with multiplicity, or, equivalently, if they generate the same characteristic polynomial:

$$\det(H_0^{(1)} - \lambda) = \det(H_0^{(2)} - \lambda) \quad \forall \lambda \in \mathbb{R}. \quad (9)$$

According to the above remarks, we will also speak of ‘isospectral systems’. Sometimes we will apply the term ‘isospectral’ to more general pairs of operators derived from $H_0^{(i)}$, $i = 1, 2$ if there is no danger of confusion. Clearly, equation (9) defines an equivalence relation \sim on \mathcal{J} . The coefficients of a characteristic polynomial $P(\lambda) = \det(H_0 - \lambda)$ can be viewed as polynomials of the coupling constants J_{xy} . These polynomials assume constant values exactly on the \sim -equivalence classes $[\mathbf{J}]_{\sim}$, which we will call *isospectral classes*. Consequently, the isospectral classes are *algebraic varieties* in \mathcal{J} , since they are defined by a finite number k of polynomial equations⁴ $p_v(\mathbf{J}) = 0$, $v = 1, \dots, k$.

In algebraic geometry there are various equivalent definitions of the *dimension* of an algebraic variety, which are, however, too technical to be reproduced here (see, for example, [10], chapter 9). For our purposes it will suffice to note that for the special case where the Jacobian matrix $(\partial p_v / \partial J_j)_{v=1 \dots k, j=1 \dots \binom{N}{2}}$ has locally a constant rank r , the corresponding isospectral classes are locally differentiable manifolds of dimension $\ell = \binom{N}{2} - r$. This is an immediate consequence of the fibration theorem (see, for example, [12], theorem 3.5.18). In particular, if $k \geq \binom{N}{2}$ and the rank of the Jacobian is maximal, $r = \binom{N}{2}$, the isospectrality classes will only consist of a discrete point set. Note, however, that the equivalence classes will never be trivial since they at least consist of the orbits of the group of discrete symmetries of \mathcal{J} generated by permutations of spin sites.

We will say that the pair (N, s) is of *isospectral type* if the corresponding space of coupling constants \mathcal{J} contains at least one isospectral equivalence class of dimension $\ell \geq 1$. The largest dimension ℓ of isospectral equivalence classes will be called the *dimension* of the isospectral type.

Functions $f : \mathcal{J} \rightarrow \mathbb{R}$ which are constant on isospectral classes will be called *isospectral invariants*.

Moreover, we will consider a special case of isospectrality in which the equivalence classes are easily calculated. Obviously, two isospectral Hamiltonians $H_0^{(1)}$ and $H_0^{(2)}$ can be related by a unitary transformation U :

$$H_0^{(2)} = U^* H_0^{(1)} U. \quad (10)$$

U maps the eigenvectors of $H_0^{(2)}$ onto the corresponding eigenvectors of $H_0^{(1)}$. In the case of a one-dimensional isospectral equivalence class parametrized by a coordinate t , U can be chosen to depend smoothly on t . Following a closed loop, the corresponding unitary transformation need not reduce to the identity transformation, but may include some phases. We will come back to this phenomenon later. If for sufficiently many curves of isospectrally equivalent points the corresponding unitary transformations $U(t)$ can, moreover, be chosen to be a one-parameter group, the system (N, s) will be called of ‘covariant isospectral’ type. More precisely, we define:

⁴ For an elementary introduction into the theory of algebraic varieties, see, e.g., [10]. Note that algebraic varieties need not be differentiable manifolds since they may contain ‘boundaries’ such as the vertex of the lightcone $x^2 + y^2 + z^2 = c^2 t^2$ or ‘hairs’ like $x = y = 0$ in the variety $(x^2 + y^2)z = 0$.

Definition 1. (N, s) is of covariant isospectral type iff it is of isospectral type and for any isospectrally equivalent $\mathbf{J}^{(1)}, \mathbf{J}^{(2)}$ which are sufficiently close, there exists an anti-Hermitian $\dim \times \dim$ -matrix Ω , and a real $\binom{N}{2} \times \binom{N}{2}$ -matrix M and some $t_0 \in \mathbb{R}$, such that for all $t \in \mathbb{R}$

$$\exp(\Omega^*t)\mathbf{J}^{(1)} \cdot \mathbf{H} \exp(\Omega t) = (\exp(-Mt)\mathbf{J}^{(1)}) \cdot \mathbf{H} \tag{11}$$

and

$$\exp(-Mt_0)\mathbf{J}^{(1)} = \mathbf{J}^{(2)}. \tag{12}$$

It follows immediately, that the orbit $\exp(-Mt)\mathbf{J}^{(1)}$ in \mathcal{J} -space corresponds to a family of isospectral Hamilton matrices. Since in our case all Hamilton matrices are real and symmetric, Ω can also be chosen as real and hence antisymmetric, and thus $\exp(\Omega t)$ represents a rotation in the real Hilbert space \mathbb{R}^{dim} .

The condition (11) may be replaced by its equivalent infinitesimal version:

$$[\mathbf{H}, \Omega] = -M^* \mathbf{H}. \tag{13}$$

This equation could be used to show that M , considered as a linear transformation in the space of $\binom{N}{2}$ -dimensional vectors with matrix entries endowed with the scalar product $\text{Tr}(\mathbf{K} \cdot \mathbf{H})$, will be an antisymmetric matrix. We will give an independent proof of this fact later.

Furthermore, we note that the set of solutions $\langle \Omega, M \rangle$ of (13) will be a Lie algebra with respect to the obvious vector and commutator operations. Hence covariant isospectral equivalence classes will be orbits of the corresponding matrix Lie groups.

Thus far we have only discussed isospectral systems in the absence of an external magnetic field. If we include the field \mathcal{H} and allow for a corresponding h -dependence of the Hamiltonian, we have to consider a slightly stronger concept of ‘complete isospectrality’.

Definition 2. Two Hamiltonians $H_0^{(1)}$ and $H_0^{(2)}$ (or, equivalently, two vectors $\mathbf{J}^{(1)}$ and $\mathbf{J}^{(2)} \in \mathcal{J}$) are called completely isospectral ($\mathbf{J}^{(1)} \approx \mathbf{J}^{(2)}$) iff for all $h \in \mathbb{R}$ $H^{(1)}(h) = H_0^{(1)} - hS^{(3)}$ and $H^{(2)}(h) = H_0^{(2)} - hS^{(3)}$ are isospectral.

Proposition 1. The following conditions are equivalent:

- (i) $\mathbf{J}^{(1)} \approx \mathbf{J}^{(2)}$;
- (ii) $P_M H_0^{(1)} P_M \sim P_M H_0^{(2)} P_M$ for all projectors P_M onto the eigenspaces of $S^{(3)}$ corresponding to the eigenvalue M ;
- (iii) $P_S H_0^{(1)} P_S \sim P_S H_0^{(2)} P_S$ for all projectors P_S onto the eigenspaces of S^2 corresponding to the eigenvalue $S(S + 1)$.

Proof.

(i) \Rightarrow (ii). Since all $H(h), h \in \mathbb{R}$, commute, there exists a system of joint eigenprojectors \mathcal{P}_{v, M_v} such that

$$H(h) = \sum_{v, M_v} (\epsilon_v - hM_v) \mathcal{P}_{v, M_v}. \tag{14}$$

Hence

$$P_M H(h) P_M = \sum_{v, M_v, M_v=M} (\epsilon_v - hM_v) \mathcal{P}_{v, M_v}. \tag{15}$$

If $H^{(1)}(h) \sim H^{(2)}(h)$ for all $h \in \mathbb{R}$, then both systems have the same set of eigenvalues $\epsilon_v - hM_v$ with the same multiplicities $\text{Tr } \mathcal{P}_{v, M_v}$. Hence, by (15), also $P_M H^{(1)}(h) P_M \sim P_M H^{(2)}(h) P_M$, in particular, $P_M H_0^{(1)} P_M \sim P_M H_0^{(2)} P_M$ for all M .

(ii) \Rightarrow (iii). The eigenspaces corresponding to \mathcal{P}_{ν, M_ν} can be further split into eigenspaces of \mathcal{S}^2 . This can be written as

$$\mathcal{P}_{\nu, M_\nu} = \sum_{S=|M_\nu|}^{S_{max}} \mathcal{P}_{\nu, M_\nu, S}. \tag{16}$$

Applying the ladder operators S^\pm gives

$$\text{Tr } \mathcal{P}_{\nu, M_\nu, S} = \text{Tr } \mathcal{P}_{\nu, S, S} = \text{Tr } \mathcal{P}_{\nu, S} \quad \text{for all } M_\nu = -S, \dots, S. \tag{17}$$

These numbers are the multiplicities of the eigenvalues of the operators $\mathbb{P}_S \mathcal{P}_M H_0^{(i)} \mathbb{P}_S \mathcal{P}_M$, which are hence isospectral for $i = 1, 2$. By summation over M we also conclude that $\mathbb{P}_S H_0^{(i)} \mathbb{P}_S$ are isospectral for $i = 1, 2$.

(iii) \Rightarrow (ii). This can be shown analogously by considering

$$\mathbb{P}_S H_0 \mathbb{P}_S = \sum_{\nu, S_\nu, S_\nu=S} \epsilon_\nu \mathcal{P}'_{\nu, S_\nu} \tag{18}$$

$$\mathcal{P}'_{\nu, S_\nu} = \sum_{M=-S_\nu}^{S_\nu} \mathcal{P}'_{\nu, S_\nu, M} \tag{19}$$

and

$$\text{Tr } \mathcal{P}'_{\nu, S_\nu} = (2S_\nu + 1) \text{Tr } \mathcal{P}'_{\nu, S_\nu, M}. \tag{20}$$

(ii) \Rightarrow (i). This follows from

$$H(h) = \sum_M (\mathcal{P}_M H_0 \mathcal{P}_M - hM). \tag{21}$$

□

Obviously, $\mathbf{J}^{(1)} \approx \mathbf{J}^{(2)}$ implies $\mathbf{J}^{(1)} \sim \mathbf{J}^{(2)}$, but not conversely: take $N = 3, s = \frac{1}{2}$, then

$$\mathbf{J}^{(1)} = \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix} \sim \mathbf{J}^{(2)} = \begin{pmatrix} -1 \\ -1 \\ -1 \end{pmatrix}$$

but $\mathbf{J}^{(1)} \not\approx \mathbf{J}^{(2)}$. Unfortunately we do not know of less trivial counter-examples. The problem is the following: being rotationally symmetric, H_0 commutes with \mathcal{S}^2 and $\mathcal{S}^{(3)}$, hence each eigenspace of H_0 with eigenvalue ϵ_ν is spanned by simultaneous eigenvectors of \mathcal{S}^2 and $\mathcal{S}^{(3)}$, say, $|v, \lambda, \mu\rangle, v = 0, \dots, D, \lambda = 1, \dots, d_\nu, \mu = -S_{v,\lambda}, \dots, S_{v,\lambda}$, such that

$$\mathcal{S}^2 |v, \lambda, \mu\rangle = S_{v,\lambda}(S_{v,\lambda} + 1) |v, \lambda, \mu\rangle \quad \mathcal{S}^{(3)} |v, \lambda, \mu\rangle = \mu |v, \lambda, \mu\rangle. \tag{22}$$

The degeneracy of the eigenvalues ϵ_ν will be

$$n_\nu = \sum_{\lambda=1}^{d_\nu} (2S_{v,\lambda} + 1). \tag{23}$$

We do not assume that the $S_{v,\lambda}$ have different values. The corresponding eigenvalues of $H(h) = H_0 - h\mathcal{S}^{(3)}$ are

$$E_{v,\lambda,\mu} = \epsilon_\nu - h\mu. \tag{24}$$

For ‘generic’ $\mathbf{J} \in \mathcal{J}$ we expect that H_0 will have no further degeneracy besides that dictated by rotational symmetry (‘minimal’ degeneracy), i.e. we expect that $d_\nu = 1$. So λ can be skipped and $\mu = -S_\nu, \dots, S_\nu$. In this case, ‘isospectrality’ and ‘complete isospectrality’ would be equivalent. So, heuristically, we may consider these two notions as having equal meaning, although we have to distinguish between them for the sake of mathematical rigour.

For the most important case of isospectral systems which are obtained by continuously varying the coupling constants we can show the following:

Proposition 2. *Any two systems joined by a continuous curve of isospectral systems are completely isospectral.*

Proof. Consider a curve $t \mapsto \mathbf{J}(t)$ and consider

$$H_0(t) \equiv \mathbf{H} \cdot \mathbf{J}(t) = \sum_\nu \epsilon_\nu \mathcal{P}_\nu(t) \tag{25}$$

and

$$\mathbb{P}_S H_0(t) \mathbb{P}_S = \sum_\nu \epsilon_\nu \mathcal{P}_\nu(t) \mathbb{P}_S. \tag{26}$$

Since $t \mapsto \mathcal{P}_\nu(t)$ is continuous and $\text{Tr}(\mathcal{P}_\nu(t) \mathbb{P}_S)$ assumes only non-negative integer values, the latter must be constant with respect to the parameter t . However, $\text{Tr}(\mathcal{P}_\nu(t) \mathbb{P}_S)$ equals the multiplicity of the eigenvalue ϵ_ν in (26). Thus all $\mathbb{P}_S H_0(t) \mathbb{P}_S, t \in \mathbb{R}$, are isospectral, and, by proposition 1, all $H_0(t)$ are completely isospectral. \square

Note that, according to this proposition, in the above counter-example the two systems cannot lie in the same connected component of an isospectral class.

We add some definitions concerning symmetrical polynomials which will be of later use: the equation

$$\prod_{n=1}^d (x + x_n) = \sum_{\nu=0}^d s_\nu x^{d-\nu} \tag{27}$$

defines the elementary symmetrical polynomials

$$s_1 = \sum_{n=1}^d x_n, \quad s_2 = \sum_{n < m} x_n x_m, \quad \dots, \quad s_d = \prod_{n=1}^d x_n. \tag{28}$$

These also appear, up to a sign, as the coefficients of the characteristic polynomial of H_0

$$p(\lambda) = \prod_{i=1}^d (\lambda - E_i) \tag{29}$$

written as polynomials of the E_i , where $d = \text{dim}$. Every other symmetric polynomial can be written uniquely as a polynomial of the s_ν (see, for example [10], section 7.1, theorem 3). This holds especially for

$$t_n = \text{Tr}(H_0^n) = \sum_{i=1}^d E_i^n. \tag{30}$$

Conversely, each s_ν can uniquely be written as a polynomial of the $t_n, n = 1 \dots d$ (see, for example [10], section 7.1, theorem 8), e.g.

$$s_5 = \frac{1}{5!} (t_1^5 - 10t_1^3 t_2 + 15t_1 t_2^2 + 20t_1^2 t_3 - 20t_2 t_3 - 30t_1 t_4 + 24t_5). \tag{31}$$

This representation is independent of the dimension d .

3. Specific heat and magnetic susceptibility

As mentioned in the introduction (completely) isospectral spin systems will give rise to the same thermodynamic functions such as specific heat and magnetic susceptibility. In this section we will state this more precisely and also prove the converse, up to the subtle distinction between complete and plain isospectrality.

Lemma 1. *Two spin systems are isospectral iff $\mathcal{T}(H_0^{(1)}) = \mathcal{T}(H_0^{(2)})$.*

Proof. Recall that $\mathcal{T}(H_0)$ was defined as the set of traces $t_n = \text{Tr}(H_0^n)$, $n = 0, \dots, \dim$. Hence the ‘only if’ part is obvious. From the remarks at the end of section 2 it follows that if two systems possess the same t_n , $n = 0, \dots, \dim$, they share also the same values of the standard symmetric polynomials $s_\nu(\mathbf{E})$ and hence have the same characteristic polynomial $p(\lambda)$. \square

Proposition 3. *Two spin systems are isospectral iff they possess the same specific heat function.*

Proof. $c(\beta)$ can be expanded into a Taylor series at $\beta = 0$:

$$c(\beta) = \left(-\frac{t_1^2}{t_0^2} + \frac{t_2}{t_0}\right)\beta^2 + \left(\frac{t_1 t_2}{t_0^2} - \frac{t_1}{t_0} \left(\frac{t_1^2}{t_0^2} - \frac{t_2}{2t_0}\right) - \frac{t_3}{2t_0}\right)\beta^3 + \dots \quad (32)$$

This is the starting point of the so-called ‘moment expansion method’ (see, e.g., [11], section 7.3). Obviously, each coefficient of β^n uniquely determines t_n , if the other traces t_m , $m < n$, are already known. Note that $t_0 = \dim$, $t_1 = 0$. Together with lemma 1 this completes the proof. \square

Now we consider again $H(h) = H_0 - hS^{(3)}$ with eigenvectors $|\nu, \lambda, \mu\rangle$ according to the previous section. By its very definition, two completely isospectral systems share the same partition function $\mathcal{Z}(\beta, h)$ and any other thermodynamic function which can be derived from it. In particular, the following holds:

Proposition 4. *Two completely isospectral systems possess the same magnetic susceptibility function.*

To tackle the converse problem, we consider $\mathcal{Z}(\beta, 0) = \sum_\nu n_\nu \exp(-\beta\epsilon_\nu)$ and define the coefficients σ_ν implicitly by

$$\text{Tr}(S^{(3)2} \exp(-\beta H_0)) = \sum_\nu \sigma_\nu n_\nu \exp(-\beta\epsilon_\nu). \quad (33)$$

In the case of minimal degeneracy, i.e. $d_\nu = 1$, we have $n_\nu = 2S_\nu + 1$ and

$$\sigma_\nu = \frac{1}{2S_\nu + 1} \sum_{\mu=-S_\nu}^{S_\nu} \mu^2 = \frac{1}{3} S_\nu(S_\nu + 1). \quad (34)$$

In the general case,

$$\begin{aligned} \sigma_\nu &= \frac{1}{n_\nu} \sum_{\lambda=1}^{d_\nu} \sum_{\mu=-S_{\nu,\lambda}}^{S_{\nu,\lambda}} \mu^2 \\ &= \frac{1}{n_\nu} \sum_{\lambda=1}^{d_\nu} \frac{1}{3} S_{\nu,\lambda}(S_{\nu,\lambda} + 1)(2S_{\nu,\lambda} + 1). \end{aligned} \quad (35)$$

Proposition 5. *Two spin systems with the same susceptibility function are isospectral.*

Proof. Since

$$\chi(\beta) = \frac{\beta}{\mathcal{Z}(\beta, 0)} \text{Tr} (S^{(3)2} \exp(-\beta H_0)) \tag{36}$$

the two systems will have the same function

$$\begin{aligned} f(\beta) &\equiv \frac{\text{Tr} (\exp(-\beta H_0) S^{(3)2})}{\text{Tr} (\exp(-\beta H_0))} \\ &= \frac{\sum_{v=0}^D \sigma_v n_v \exp(-\beta \varepsilon_v)}{\sum_{v=0}^D n_v \exp(-\beta \varepsilon_v)}. \end{aligned} \tag{37}$$

Since $\varepsilon_0 < \varepsilon_1 < \varepsilon_2 < \dots$, the terms $\exp(-\beta \varepsilon_v)$ are of different orders of magnitude for $\beta \rightarrow \infty$. The first term increasingly dominates, hence

$$\lim_{\beta \rightarrow \infty} f(\beta) = \frac{\sigma_0 n_0 \exp(-\beta \varepsilon_0)}{n_0 \exp(-\beta \varepsilon_0)} = \sigma_0. \tag{38}$$

If we subtract this limit from $f(\beta)$, the dominant term becomes asymptotically

$$\begin{aligned} f(\beta) - \sigma_0 &\simeq_{\beta \rightarrow \infty} \frac{\sigma_1 n_1 \exp(-\beta \varepsilon_1)}{n_0 \exp(-\beta \varepsilon_0)} \\ &= \frac{\sigma_1 n_1}{n_0} \exp(-\beta(\varepsilon_1 - \varepsilon_0)). \end{aligned} \tag{39}$$

In the next step we have

$$f(\beta) - \sigma_0 - \frac{\sigma_1 n_1}{n_0} \exp(-\beta(\varepsilon_1 - \varepsilon_0)) \simeq_{\beta \rightarrow \infty} \frac{\sigma_2 n_2}{n_0} 2 \exp(-\beta(\varepsilon_2 - \varepsilon_0)) \tag{40}$$

and so on. In this way, from the behaviour of $f(\beta)$ for $\beta \rightarrow \infty$, we may extract the values

$$\sigma_0, \frac{\sigma_1 n_1}{n_0}, \dots, \frac{\sigma_v n_v}{n_0}, \dots, \frac{\sigma_D n_D}{n_0}$$

and

$$\varepsilon_1 - \varepsilon_0, \varepsilon_2 - \varepsilon_0, \dots, \varepsilon_v - \varepsilon_0, \dots, \varepsilon_D - \varepsilon_0.$$

Let $t_n = \text{Tr}(H_0^n)$ as above and $\mu_n \equiv \text{Tr}(H_0^n S^{(3)2})$, $n \in \mathbb{N}$. $\mu_0 = \text{Tr}(S^{(3)2})$ can be calculated independently of H_0 . Since

$$\mu_0 = \sum_{v=0}^D \sigma_v n_v = n_0 \sum_{v=0}^D \frac{\sigma_v n_v}{n_0} \tag{41}$$

n_0 and hence $\sigma_v n_v$, $v = 1, \dots, D$, are also uniquely determined.

Next we consider the Taylor expansion of $f(\beta)$ at $\beta = 0$:

$$\begin{aligned} f(\beta) &= \frac{\mu_0}{t_0} - \frac{\mu_1}{t_0} \beta + \left(-\frac{t_2 \mu_0}{2t_0^2} + \frac{\mu_2}{2t_0} \right) \beta^2 + \left(\frac{t_3 \mu_0}{6t_0^2} + \frac{t_2 \mu_1}{2t_0^2} - \frac{\mu_3}{6t_1} \right) \beta^3 \\ &\quad + \left(\mu_0 \left(\frac{t_2^2}{4t_0^3} - \frac{t_4}{24t_0^2} \right) - \frac{t_3 \mu_1}{6t_0^2} - \frac{t_2 \mu_2}{4t_0^2} + \frac{\mu_4}{24t_0} \right) \beta^4 \\ &\quad + \dots \end{aligned} \tag{42}$$

Recall that t_0, μ_0 are known. The linear term then gives

$$\mu_1 = \text{Tr}(H_0 S^{(3)2}) = \sum_{v=0}^D \sigma_v n_v \varepsilon_v. \tag{43}$$

On the other hand, we know the left-hand side of

$$\sum_{\nu=0}^D \sigma_\nu n_\nu (\varepsilon_\nu - \varepsilon_0) = \mu_1 - \varepsilon_0 \mu_0 \tag{44}$$

hence ε_0 and all $\varepsilon_\nu, \nu = 1, \dots, D$ are also known. Similarly,

$$\sum_{\nu=0}^D \sigma_\nu n_\nu (\varepsilon_\nu - \varepsilon_0)^2 = \mu_2 - 2\varepsilon_0 \mu_1 + \varepsilon_0^2 \mu_0 \tag{45}$$

hence μ_2 is known and from the β^2 -term in (42) also t_2 , and so on.

Eventually, we obtain all $t_n, \mu_n, n = 2, \dots, \dim$ solely from $\chi(\beta)$. According to lemma 1 this gives us all the eigenvalues of H_0 with multiplicity, i.e. $n_\nu, \nu = 0, \dots, D$ and the two spin systems are isospectral. \square

The proof does not give complete isospectrality: if some eigenvalue ε_ν belongs to different $S_{\nu\lambda}, \lambda > 2$ then from $n_\nu = \sum_\lambda (2S_{\nu\lambda} + 1)$ and

$$\sigma_\nu = \frac{\sum_\lambda \frac{1}{3} S_{\nu\lambda} (S_{\nu\lambda} + 1) (2S_{\nu\lambda} + 1)}{\sum_\lambda (2S_{\nu\lambda} + 1)}$$

the $S_{\nu,\lambda}$ cannot be uniquely determined.

4. Some isospectral invariants

Criteria for non-isospectrality could, in principle, be checked by brute-force methods: calculate the characteristic polynomial of the matrix $H_0 = \mathbf{J} \cdot \mathbf{H}$, say $p(\lambda) = \sum_{\nu=0}^{\dim} c_\nu \lambda^\nu$. Select $\binom{N}{2}$ different coefficients c_i, c_j, \dots ($\nu = \dim$ being excluded since $c_{\dim} = 1$) and calculate the Jacobian determinant

$$Jac(\mathbf{J}) \equiv \frac{\partial(c_i, c_j, \dots)}{\partial(J_1, \dots, J_{\binom{N}{2}})} \tag{46}$$

preferably by using computer algebra software. If the Jacobian vanishes nowhere, according to the remarks in section 2, (N, s) cannot be of isospectral type.

In practice this method will, even for small N and s , rapidly become extremely memory- and time-consuming. To simplify the problem one could—in the case of complete isospectrality—restrict oneself to subspaces that are invariant under H_0 , for example, subspaces $\mathcal{H}(M)$ of constant magnetic quantum number M .

The space with maximal $M, \mathcal{H}(M = Ns)$ is one dimensional and is spanned by the product state

$$\varphi_0 = |s, s, \dots, s\rangle \tag{47}$$

which is an eigenstate of $\mathbf{J} \cdot \mathbf{H}$ for all $\mathbf{J} \in \mathcal{J}$ with eigenvalue

$$E_0 = s^2 J \equiv s^2 \sum_{x < y} J_{xy}. \tag{48}$$

This proves the first part of the following:

Lemma 2. *If $\mathbf{J}^{(1)} \cdot \mathbf{H}$ and $\mathbf{J}^{(2)} \cdot \mathbf{H}$ are completely or covariantly isospectral, then*

$$\sum_{x < y} J_{xy}^{(1)} = \sum_{x < y} J_{xy}^{(2)} \tag{49}$$

i.e. $\mathbf{J}^{(1)}$ and $\mathbf{J}^{(2)}$ lie in the same hyperplane perpendicular to $\mathbf{1} \equiv (1, 1, \dots, 1)$.

Proof. If all $J_{xy} \geq 0$, then $E_0 = s^2 J$ will be the maximal eigenvalue of $\mathbf{J}^{(2)} \cdot \mathbf{H}$. In fact, $\langle \varphi_0 | H_{xy} \varphi_0 \rangle$ is the maximal expectation value for each H_{xy} . Hence J is an isospectral invariant at least in the domain $\mathcal{J}^+ \equiv \{J | \text{all } J_{xy} \geq 0\}$. Now assume covariant isospectrality and let $t \mapsto \mathbf{J}(t) = \exp(-Mt)\mathbf{J}(0)$ be an isospectral curve which will be restricted to \mathcal{J}^+ . According to what has been said before, $0 = \dot{J} = \frac{d}{dt} \langle J | \mathbf{1} \rangle = \langle \dot{J} | \mathbf{1} \rangle = \langle -M\mathbf{J} | \mathbf{1} \rangle = \langle J | -M^* \mathbf{1} \rangle = \langle J | M\mathbf{1} \rangle$ for all $J \in \mathcal{J}^+$. Since \mathcal{J}^+ linearly generates \mathcal{J} it follows that $M\mathbf{1} = \mathbf{0}$, i.e. all row sums of M vanish and J is an isospectral invariant on the whole space \mathcal{J} . \square

Before proceeding with $M = Ns - 1$ we will show that also

Lemma 3. $\|J\|^2 = \sum_{x < y} J_{xy}^2$ is an isospectral invariant.

Proof. Obviously, $\text{Tr}(H_0^2)$, the sum of all eigenvalues squared, is the same for isospectral Hamiltonians. After expanding the square $(\mathbf{J} \cdot \mathbf{H})^2$ one realizes that only those products $H_{xy}H_{uv}$ have a non-zero trace where $x = u$ and $y = v$. Hence $\text{Tr}(H_0^2) = \sum_{x < y} J_{xy}^2 \text{Tr}(H_{xy}^2) = (\sum_{x < y} J_{xy}^2) \cdot \frac{1}{3}s^2(s+1)^2(2s+1)^N$ (see appendix A). The actual value for $\text{Tr}(H_{xy}^2)$ is irrelevant for the proof; what matters only is that it is independent of x, y . This concludes the proof. \square

From lemma 3 it follows immediately that in the covariant isospectral case the matrix $\exp(-tM)$ leaves the Euclidean norm $\|\cdot\|$ invariant and thus must be an orthogonal transformation and its generator $-M$ will be antisymmetric.

5. The triangle ($N = 3$)

5.1. $s = \frac{1}{2}$

Next, we consider eigenvalues with eigenvectors in the subspace $\mathcal{H}(M = Ns - 1)$, but restricted to the case $N = 3$. With the abbreviations

$$J = J_{12} + J_{23} + J_{13} \tag{50}$$

$$\Gamma = J_{12}J_{23} + J_{12}J_{13} + J_{23}J_{13} \tag{51}$$

the eigenvalues are calculated to be (see [13], section 62, exercise 2)

$$E_0 = s^2 J \quad E_{1,2} = s(s-1)J \pm \sqrt{J^2 - 3\Gamma}. \tag{52}$$

Hence the first three eigenvalues are constant on curves with constant J and Γ . These are circles with radius

$$r = \sqrt{\frac{2}{3}J^2 - 2\Gamma} \tag{53}$$

the centre of which is located on the line $J_{12} = J_{23} = J_{13}$, including the degenerate case $r = 0$.

For $s = \frac{1}{2}$ the list of eigenvalues is already exhausted: due to rotational symmetry the value E_0 is four-fold degenerate ($S = \frac{3}{2}$) and the $E_{1,2}$ are twofold degenerate ($S = \frac{1}{2}$). We conclude:

Proposition 6. *The system $N = 3, s = \frac{1}{2}$ is of complete isospectral type with dimension two.*

We now consider the question of whether the triangle with $s = \frac{1}{2}$ is of covariant isospectral type, i.e. we seek solutions of

$$[H, \Omega] = MH \tag{54}$$

where Ω and M are antisymmetric. Let $T = T^3$ be the unitary left shift operator which represents a cyclic permutation of the spin sites. Then a solution of (54) is given by

$$\Omega = \frac{1}{2\sqrt{3}}(T - T^*) \quad M = \frac{1}{\sqrt{3}} \begin{pmatrix} 0 & 1 & -1 \\ -1 & 0 & 1 \\ 1 & -1 & 0 \end{pmatrix}. \quad (55)$$

Ω can also be written as

$$\Omega = \frac{i}{4\sqrt{3}} \sigma_3 \cdot (\sigma_1 \times \sigma_2) \quad (56)$$

where the σ_i ($i = 1, 2, 3$) denote the Pauli matrices. Obviously, Ω is rotationally symmetric which entails complete isospectrality. The factor $\frac{1}{\sqrt{3}}$ is chosen such that the parameter t in $\exp(tM)$ will be just the angle of rotation. $T^3 = T$ entails $\Omega^3 = -\frac{1}{4}\Omega$, hence the exponential series of $\exp(t\Omega)$ will be actually a polynomial in Ω :

$$\exp(t\Omega) = 1 + 2\Omega \sin \frac{t}{2} + (2\Omega)^2 \left(1 - \cos \frac{t}{2}\right). \quad (57)$$

For special values of t we obtain

$$\exp\left(\frac{4\pi}{3}\Omega\right) = T \quad (58)$$

$$\exp(4\pi\Omega) = T^3 = \mathbb{1} \quad (59)$$

$$\exp(2\pi\Omega) = \frac{2}{3}(T^2 + T - \frac{1}{2}). \quad (60)$$

The last expression can be rewritten using

$$\tilde{H} \equiv \begin{pmatrix} 1 \\ 1 \\ 1 \end{pmatrix} \cdot \mathbf{H} = \frac{3}{4} (\mathbb{P}_{3/2} - \mathbb{P}_{1/2}) \quad (61)$$

where $\mathbb{P}_{3/2}$ (respectively, $\mathbb{P}_{1/2}$) denotes the projector onto the subspace $S = \frac{3}{2}$ (respectively, $S = \frac{1}{2}$). The result is

$$\exp(2\pi\Omega) = \mathbb{P}_{3/2} - \mathbb{P}_{1/2}. \quad (62)$$

This means that an eigenstate of H_0 with $S = \frac{1}{2}$ acquires a phase of π after a full rotation in \mathcal{J} -space, analogous to the occurrence of Berry phases for adiabatic loops in parameter space. Summarizing, we state the following proposition which, in essence, is due originally to Grachev [6]:

Proposition 7. *The system $N = 3, s = \frac{1}{2}$ is of both completely and covariantly isospectral type.*

5.2. A physical example

An interesting and timely application of this theory is provided by the example of the molecular magnet $(\text{CN}_3\text{H}_6)_4\text{Na}_2[\text{H}_4\text{V}_6\text{P}_4\text{O}_{30}(\text{CH}_2)_3\text{CCH}_2\text{OH}_2] \cdot 14 \text{H}_2\text{O}$ which features two uncoupled systems of three V^{4+} ($s = \frac{1}{2}$) ions that interact via antiferromagnetic Heisenberg exchange. It has been proposed [3] that the Coulomb interaction between an Na ion and two of the three V^{4+} ions gives rise to what is essentially an isosceles triangle, with the distances between the three vanadium ions being 3.20, 3.21 and 3.36 Å. It is then quite reasonable to assume that the three

exchange constants satisfy $J_{12} = J_{13} \neq J_{23}$. In fact, calculation of the weak-field susceptibility has yielded results that are in excellent agreement with accurate susceptibility measurements from room temperature down to 2 K upon assigning the values $J_{12} = J_{13} = 64.7$ K and⁵ $J_{23} = 7.5$ K [1, 2]. Moreover, the calculated energy level spacings that follow from these assignments have recently been confirmed to good accuracy in a direct manner by inelastic neutron scattering [14]. Nevertheless, as the work of this section has shown, the identical energy levels and the identical temperature-dependent susceptibility emerge for the continuous choices of the three different exchange constants that lie on curves with $J = 136.9$ K and $\Gamma = 5156.6$ K².

5.3. $s > \frac{1}{2}$

For $s > \frac{1}{2}$ we consider the next subspace $\mathcal{H}(M = 3s - 2)$. The characteristic polynomial $p(\lambda) = \sum_{v=1}^6 c_v \lambda^v$ has been calculated using MATHEMATICA 4.0, but is too complicated to be presented here. One particular Jacobian reads

$$\frac{\partial(c_3, c_4, c_5)}{\partial(J_{12}, J_{23}, J_{13})} = (3s - 1)(6s - 1)(1 - 8s + 6s^2)(1 - 6s + 15s^2). \quad (63)$$

This is a polynomial in s which has no integer or half-integer roots. Therefore, we have proved the following:

Proposition 8. *The system $N = 3$, $s > \frac{1}{2}$ is not of complete isospectral type.*

6. Isospectrality for $N > 3$

The question arises as to whether our result that complete isospectrality only occurs for $s = \frac{1}{2}$ also holds for $N > 3$. Our method of calculating the Jacobian (46) for arbitrary s will no longer work for larger N . However, we can prove a weaker statement, namely:

Proposition 9. *Systems with $N > 3$ cannot be of covariantly isospectral type.*

Proof. For this we need a trace formula which will be explained in appendix A:

$$\text{Tr}(H_0^3) = \sum_{x < y} (J_{xy})^3 \left(-\frac{1}{6}s^2(s+1)^2(2s+1)^N\right) + \sum_{x < y < z} J_{xy}J_{yz}J_{xz} \left(\frac{2}{3}s^3(s+1)^3(2s+1)^N\right). \quad (64)$$

From the isospectral invariance of $\text{Tr}(H^3)$ we also conclude that

$$f_3 \equiv \sum_{x < y} (J_{xy})^3 - 4s(s+1) \sum_{x < y < z} J_{xy}J_{yz}J_{xz} \quad (65)$$

will be invariant. Now we consider four different spin sites (using $N > 3$) denoted by 1, 2, 3, 4 and consider vectors $\mathbf{J} \in \mathcal{J}$ which have vanishing components except possibly for $J_{12}, J_{13}, J_{23}, J_{14}, J_{24}, J_{34}$. One-parameter isospectral curves passing through \mathbf{J} satisfy

$$\frac{d}{dt} f_3(\mathbf{J}(t)) = 0. \quad (66)$$

Using (65) and $\frac{d}{dt} \mathbf{J}(t) = -M\mathbf{J}(t)$, equation (66) can be written as an equation which is linear with respect to the 15 relevant matrix entries of M and trilinear with respect to the six

⁵ It is usual to measure the coupling constants in units of Kelvin. The corresponding energies are obtained by multiplying with the Boltzmann constant k_B .

non-vanishing components of \mathbf{J} . Using MATHEMATICA 4.0 it is easy to show that (66) only has the trivial solution $M = 0$. For example, one may randomly choose 15 vectors \mathbf{J} (it suffices to consider components $-1, 0, 1$) and cast the corresponding 15 equations of the form (66) into matrix form. Non-trivial solutions exist only if the determinant of this matrix, which is a polynomial in s of degree 30 with integer coefficients, vanishes. However, the zeros of the polynomial can be computed numerically and shown not to attain half-integer or integer values. \square

So it seems that the concept of covariant isospectrality is of little use, having only one single application for $N = 3, s = \frac{1}{2}$. However, covariance may be restored for $N \geq 4$ if the class of admissible Hamiltonians is suitably extended, e.g. to also include Hamiltonians which are bi-quadratic in the spin observables. However, this is beyond the scope of the present paper.

Of course, our proposition 9 does not exclude plain isospectrality for $N > 3$. Indeed, we will show that the system $N = 4, s = \frac{1}{2}$ is completely isospectral, albeit not covariantly isospectral, in the next section.

7. The $s = \frac{1}{2}$ tetrahedron case

In the case $N = 4, s = \frac{1}{2}$ it is still possible to calculate the coefficients of the characteristic polynomials of H_0 restricted to the subspaces with $M = 0, S = 2, 1, 0$. Obviously, this is enough in order to study complete isospectrality since all eigenvalues of H_0 appear within these subspaces. It turns out that all coefficients can be written as functions of four fundamental invariants I_1, I_2, I_3, I_4 . These can most conveniently be written in terms of new coordinates in \mathcal{J} , which are defined as half the sums and differences of the coupling constants of adjacent edges:

$$S_{12} \equiv \frac{1}{2}(J_{12} + J_{34}) \quad D_{12} \equiv \frac{1}{2}(J_{12} - J_{34}) \quad (67)$$

etc.

Proposition 10. *Two spin systems with coupling constants $\mathbf{J}^{(1)}, \mathbf{J}^{(2)}$ with $N = 4, s = \frac{1}{2}$ are completely isospectral iff the following four functions assume the same values for $\mathbf{J}^{(1)}$ and $\mathbf{J}^{(2)}$:*

$$I_1 = D_{12}^2 + D_{13}^2 + D_{14}^2 \quad (68)$$

$$I_2 = S_{12}^2 + S_{13}^2 + S_{14}^2 \quad (69)$$

$$I_3 = S_{12} + S_{13} + S_{14} \quad (70)$$

$$I_4 = 2D_{12}D_{13}D_{14} + D_{12}^2S_{13} + D_{13}^2S_{12} + D_{14}^2S_{13} - S_{12}S_{13}S_{14}. \quad (71)$$

Now let I' be the functional matrix obtained by partial differentiation of I_1, I_2, I_3, I_4 with respect to its six arguments S_{12}, \dots, D_{14} . The rank of I' assumes its maximal value of 4 iff no determinant of the 15 possible 4×4 submatrices of I' vanishes. We denote the subset of those points with maximal rank by $\mathcal{R} \subset \mathcal{J}$. If an isospectral class lies entirely within \mathcal{R} it will be a two-dimensional submanifold of the six-dimensional space \mathcal{J} . This follows by a well known theorem of differential geometry (see, e.g., [12], theorem 3.5.4). We will call this case *generic* and the other cases *exceptional*.

Although \mathcal{J} is six dimensional, one can visualize the two-dimensional submanifolds in the generic case. $I_1 = \text{constant}$ defines a sphere in the three-dimensional D -space with coordinates D_{12}, D_{13}, D_{14} . $I_2 = \text{constant}$ and $I_3 = \text{constant}$ define the intersection of a

sphere and a plane, i.e. a circle in \mathcal{S} -space. For given D the last equation $I_4(D, \mathcal{S}) = \text{constant}$ picks out a finite number (actually ≤ 6) of points in that circle. If the corresponding angles ψ_ν are drawn as different radii $r_\nu = 1 + \frac{\psi_\nu}{4\pi}$ in D -space (identifying points with $r_\nu = \frac{1}{2}$ and $\frac{3}{2}$) we obtain a surface folded in a complicated way.

A large number of two-dimensional generic isospectral classes have been identified numerically. The exceptional classes are one- or zero-dimensional and will be discussed further in appendix B.

8. Heuristic arguments for the (non-)occurrence of isospectrality

In the previous sections we have studied isospectrality for the cases $s = \frac{1}{2}, N = 3, 4$ and excluded certain other cases, e.g. complete isospectrality for $s > \frac{1}{2}$ and covariant isospectrality for $N > 3$. However, we have been unable to present a complete list of criteria for the (non-)occurrence of isospectrality. What is also missing is some simple and intuitive argument as to why isospectrality is so rare. As a substitute for a complete theory we will, in this section, provide some heuristic arguments for the (non-)occurrence of isospectrality which may also give more insight into isospectrality than detailed proofs. We think that these arguments could be made rigorous as far as necessary conditions for isospectrality in the cases $s = \frac{1}{2}, N = 3, 4, 5$ are involved. (See the remarks in section 2 on the dimension of isospectral classes and the fibration theorem.) However, a detailed proof would require technical issues from the theory of algebraic varieties which are beyond the scope of this paper and, moreover, would appear as superfluous given that isospectrality in some of these cases has already been proven by case inspection.

The heuristic argument goes as follows: isospectrality will (only) occur if the number $\binom{N}{2}$ of bonds between spin pairs exceeds the number L of independent eigenvalues of H_0 . In this case, typically the systems corresponding to an $(n = \binom{N}{2} - L)$ -dimensional sub-variety of \mathcal{J} will possess the same eigenvalues. The argument may even be applied if one is not aware of all relations among the eigenvalues which determine L . In such a case of unknown relations one would perhaps overestimate L and hence underestimate the dimension n of the isospectrality classes but, depending on the case, one could correctly predict the occurrence of isospectrality.

When counting the number L of independent eigenvalues one first has to consider the $(2S + 1)$ -fold degeneracy dictated by the rotational invariance of H_0 . In the simplest example, one has to couple $N = 2$ spins $s = \frac{1}{2}$, obtaining one triplet and one singlet as eigenspaces of H_0 , symbolically $2 \times 2 = 3 + 1$. Thus there are not four, but only two independent eigenvalues of H_0 . Similarly, for $N = 3$ one has $2 \times 2 \times 2 = 4 + 2 + 2$, hence three independent eigenvalues. In the latter case there are $\binom{3}{2} = 3$ bonds. Thus the heuristic argument does not yet explain isospectrality with $(n = 1)$ -dimensional classes. However, it is easy to find a ‘missing relation’ among the eigenvalues which reduces the number of independent eigenvalues to $L = 2$: it is just the relation $\text{Tr } H_0 = 0$ which yields a linear relation of the form $4E_1 + 2E_2 + 2E_3 = 0$ between the eigenvalues E_ν .

For $s = \frac{1}{2}$ and arbitrary N it can be shown that there are exactly $\binom{N}{\lfloor N/2 \rfloor}$ independent eigenvalues due to rotational degeneracy and hence, considering $\text{Tr } H_0 = 0, L \leq \binom{N}{\lfloor N/2 \rfloor} - 1$. In this manner we obtain the results summarized in table 1.

For large N the entries in the second column of table 1 grow asymptotically as $2^N \sqrt{\frac{2}{\pi N}}$, and hence almost exponentially, whereas $\binom{N}{2}$ grows only quadratically. Therefore, our heuristic argument will only predict isospectrality in the cases $s = \frac{1}{2}, N = 3, 4, 5$ but not for larger N . For larger $s > \frac{1}{2}$ the growth of the second column will prevail from the outset (see table 2) and

Table 1. Occurrence of isospectrality for $s = \frac{1}{2}$ and arbitrary N based on a heuristic argument on the number of bonds (third column) and the maximal number of independent eigenvalues (second column).

N	$\binom{N}{\lfloor N/2 \rfloor} - 1$	$\binom{N}{2}$	Isospectrality expected
2	1	1	No
3	2	3	Yes
4	5	6	Yes
5	9	10	Yes
6	19	15	No
7	34	21	No
\vdots	\vdots	\vdots	No

Table 2. Non-occurrence of isospectrality for $s = 1$ and small N based on a heuristic argument on the number of bonds (third column) and the maximal number of independent eigenvalues (second column).

N	$L \leq$	$\binom{N}{2}$	Isospectrality expected
2	3	1	No
3	8	3	No
4	24	6	No
\vdots	\vdots	\vdots	No

our argument will not even be applicable for small N . Of course, this does not strictly exclude isospectrality for those cases, but makes it very unlikely in our opinion.

There is another aspect which shows up in table 1 and which we now discuss: note that for $N = 4$ the difference between the numbers in the third and the second column, $6 - 5 = 1$, would only explain one-dimensional isospectrality classes, whereas we have encountered two-dimensional classes in section 7. This indicates that the correct L should be 4, not 5, and that there is a further ‘missing relation’ between the eigenvalues of H_0 , comparable to $\text{Tr } H_0 = 0$. Indeed, as shown in the following paragraph, there holds a general property of the eigenvalues of arbitrary Heisenberg Hamiltonians H_0 corresponding to the distribution of the eigenvalues among the quantum numbers S .

As above, denote by \mathbb{P}_S the projector onto the eigenspace of S^2 with the eigenvalue $S(S+1)$. Then

$$\text{Tr}(H_0 \mathbb{P}_S) = \sum_{x < y} J_{xy} \text{Tr}(\mathcal{S}_x \cdot \mathcal{S}_y \mathbb{P}_S). \quad (72)$$

Since S^2 and hence all \mathbb{P}_S commute with arbitrary permutations of spin sites, the last factor in (72) does not depend on x, y and can be factored out:

$$\text{Tr}(H_0 \mathbb{P}_S) = \left(\sum_{x < y} J_{xy} \right) \text{Tr}(\mathcal{S}_1 \cdot \mathcal{S}_2 \mathbb{P}_S) \quad (73)$$

$$= J \text{Tr}(\mathcal{S}_1 \cdot \mathcal{S}_2 \mathbb{P}_S). \quad (74)$$

Being independent of the J_{xy} this factor can be calculated for any suitable H_0 , e.g. the one with constant $J_{xy} \equiv 1$,

$$\tilde{H}_0 = \frac{1}{2} (S^2 - Ns(s+1)) \quad (75)$$

which yields, after some computation,

$$\text{Tr}(H_0 \mathbb{P}_S) = J \frac{1}{N(N-1)} (S(S+1) - Ns(s+1)) \text{Tr} \mathbb{P}_S. \tag{76}$$

Hence for all H_0 , the vectors with the components $(\text{Tr}(H_0 \mathbb{P}_S))_{S=S_{min}, \dots, S_{max}}$ are proportional to the constant vector given by the right-hand side of (76) with $J = 1$. This gives a number of $S_{max} - S_{min} = \lfloor Ns \rfloor$ independent linear equations for the eigenvalues of H_0 . Of course, $\text{Tr} H_0 = \sum_S \text{Tr}(H_0 \mathbb{P}_S) = 0$ is a consequence of these equations and must not be counted separately. For $s = \frac{1}{2}$, $N = 4, 5$ we obtain $\lfloor Ns \rfloor = 2$ independent equations, which explains the two-dimensional classes for $N = 4$ we found in section 7, and predicts, at least, two-dimensional classes for the case $N = 5$ not yet analysed in detail. If this case would show isospectrality classes of dimension $n > 2$ one could try to explain this by invoking more complicated relations derived from the higher moments, $\text{Tr}(H_0^k \mathbb{P}_S)$, $k > 1$. However, we will not further pursue this question here.

9. Conclusion

We summarize our results in table 3. It is in order to add some remarks on the possibility of determining the coupling constants J . Our results on the limits of uniquely determining the values of J in the case of isospectrality does not mean that these values could not be determined otherwise. First, we did not consider thermodynamical functions which do not come solely from the partition function, such as correlation functions, etc. Second, we do not adhere to a positivistic attitude which would, in principle, deny the physical reality of unmeasurable quantities. As in other domains of physics, these parameters could also be determined with the aid of additional assumptions, e.g. based on the symmetry of the molecules and supported by chemical considerations, which although plausible have not been confirmed directly. So we think that the situation is different from theories with gauge freedom.

Table 3. Our results for the occurrence of various types of isospectrality for different N, s . The proven results are indicated as ‘yes’ or ‘no’.

N	s	Plain isospectrality	Complete isospectrality	Covariant isospectrality
3	$\frac{1}{2}$	Yes	Yes	Yes
3	$> \frac{1}{2}$	Unlikely	No	Unlikely
4	$\frac{1}{2}$	Yes	Yes	No
4	$> \frac{1}{2}$	Unlikely	No	No
5	$\frac{1}{2}$	Likely	Likely	No
> 5	$\frac{1}{2}$	Unlikely	Unlikely	No
> 5	$> \frac{1}{2}$	Unlikely	No	No

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Appendix A. Trace formulae

Expanding the terms occurring in the trace

$$\text{Tr } H_0^n = \text{Tr} \left(\sum_{x < y} J_{xy} \sum_{i=1}^3 S_x^{(i)} S_y^{(i)} \right)^n \tag{A1}$$

and using $\text{Tr}(A \otimes B) = (\text{Tr } A)(\text{Tr } B)$ one ends up with terms of the form

$$\text{Tr}(A_1 \dots A_\ell), A_v \in \{S^{(1)}, S^{(2)}, S^{(3)}\}. \tag{A2}$$

Here the spin operators without a site index denote operators operating in the single site Hilbert space \mathbb{C}^{2s+1} , not total spin operators.

Let $\ell_i (i = 1, 2, 3)$ denote the number of occurrences of $S^{(i)}$ in the product $A_1 \dots A_\ell$. One can easily show that $\text{Tr}(A_1 \dots A_\ell)$ is non-zero only if all ℓ_i are even or all ℓ_i are odd. We give a list of the simplest cases, where the trace is non-zero:

$$\ell = 0: \quad \text{Tr}(\mathbb{1}) = 2s + 1 \tag{A3}$$

$$\ell = 2: \quad \text{Tr } S^{(i)2} = \sum_{m=-s}^s m^2 = \frac{1}{3}s(s+1)(2s+1) \tag{A4}$$

$$\ell = 3: \quad \text{Tr } S^{(1)} S^{(2)} S^{(3)} = -\text{Tr } S^{(3)} S^{(2)} S^{(1)} = \frac{i}{6}s(s+1)(2s+1). \tag{A5}$$

From this we obtain for $n = 2$:

$$\text{Tr } H_0^2 = \sum_{x < y} J_{xy}^2 \sum_{i=1}^3 \text{Tr} (S_x^{(i)2} \otimes S_y^{(i)2} \otimes \mathbb{1}_{N-2}) \tag{A6}$$

$$= \left(\sum_{x < y} J_{xy}^2 \right) \frac{1}{3}s^2(s+1)^2(2s+1)^N. \tag{A7}$$

For $n = 3$ there occur two kinds of non-zero terms:

$$\text{Tr} (S_x^{(1)} S_x^{(2)} S_x^{(3)} \otimes S_y^{(1)} S_y^{(2)} S_y^{(3)} \otimes \mathbb{1}_{N-2}) \tag{A8}$$

and those terms obtained by permutations of $\{1, 2, 3\}$, and

$$\text{Tr} (S_x^{(i)2} \otimes S_y^{(i)2} \otimes S_z^{(i)2} \otimes \mathbb{1}_{N-3}) \tag{A9}$$

where x, y, z are pairwise distinct and $i = 1, 2, 3$. Consequently, we obtain

$$\text{Tr } H_0^3 = T_1 + T_2 \tag{A10}$$

$$T_1 = \sum_{x < y} J_{xy}^3 3!(2s+1)^{N-2} (\text{Tr } S^{(1)} S^{(2)} S^{(3)})^2 \tag{A11}$$

$$= -\sum_{x < y} J_{xy}^3 \frac{1}{6}s^2(s+1)^2(2s+1)^N \tag{A12}$$

$$T_2 = 3! \left(\sum_{x < y < z} J_{xy} J_{xz} J_{yz} \right) (2s+1)^{N-3} \sum_{i=1}^3 (\text{Tr } S^{(i)2})^3 \tag{A13}$$

$$= \left(\sum_{x < y < z} J_{xy} J_{xz} J_{yz} \right) \frac{2}{3}s^3(s+1)^3(2s+1)^N. \tag{A14}$$

Appendix B. The exceptional cases

Here we collect some properties of isospectral classes for $N = 4, s = \frac{1}{2}$ which belong to the exceptional case. Although we did not obtain a complete classification of all isospectral classes these results may be useful for further studies.

The subset $\mathcal{J} \setminus \mathcal{R}$, which denotes the complement of \mathcal{R} , is characterized by the vanishing of all 4×4 submatrices of I' , hence it will also be an algebraic variety. After some computations one shows that $\mathcal{J} \setminus \mathcal{R}$ is a union $\mathcal{J} \setminus \mathcal{R} = \mathcal{C}_1 \cup \mathcal{C}_2 \cup \mathcal{C}_3$ of three simpler varieties given by the following equations:

$$\mathcal{C}_1: I_1 = D_{12}^2 + D_{13}^2 + D_{14}^2 = 0 \quad (\text{B1})$$

$$\mathcal{C}_2: I_2 = 3I_3^2 \quad \text{i.e.} \quad S_{12} = S_{13} = S_{14} \quad (\text{B2})$$

$$\mathcal{C}_3: f = g = 0 \quad (\text{B3})$$

where

$$f \equiv (D_{12}^2 - D_{14}^2)D_{13} + D_{12}D_{14}(S_{14} - S_{12}) = 0 \quad (\text{B4})$$

$$g \equiv (D_{13}^2 - D_{14}^2)D_{12} + D_{13}D_{14}(S_{14} - S_{13}) = 0. \quad (\text{B5})$$

In the first case it is clear that an isospectral class lies entirely inside \mathcal{C}_1 or outside \mathcal{C}_1 , since I_1 is constant on this class. An analogous remark applies for \mathcal{C}_2 but the case \mathcal{C}_3 is more complicated. In the first case with $I_1 = 0$ the isospectral classes are zero dimensional, since the circles defined by $I_2 = i_2$ and $I_3 = i_3$ intersect the variety $I_4 = -S_{12}S_{13}S_{14} = i_4$ at most at six points. In the second case with $S_{12} = S_{13} = S_{14} = \sigma$ the isospectral classes are the one-dimensional intersections of the sphere $I_1 = i_1, i_1 > 0$ and the variety $I_4 = 2D_{12}D_{13}D_{14} + (D_{12}^2 + D_{13}^2 + D_{14}^2)\sigma - \sigma^3 = i_4$. In the third case we have shown, using the eliminate command of MATHEMATICA, that $f = g = 0$ implies an equation of the form $P(I_1, I_2, I_3, I_4) = 0$, namely

$$P = -108I_4^2 + 4I_3I_4(9I_1 + 18I_2 - 5I_3^2) + (I_1 + 2I_2 - I_3^2)^2(2I_2 + 4I_2 - I_3^2). \quad (\text{B6})$$

Hence those isospectral classes with $P(I_1, I_2, I_3, I_4) \neq 0, I_1 \neq 0, I_2 \neq 3I_3^2$ lie entirely inside \mathcal{R} and belong to the generic case. We conjecture that also the converse holds, namely that $P = 0$ implies $f = g = 0$ but could not prove it. By calculating the corresponding Groebner bases, it can be shown that P and f, g generate different ideals in the ring of polynomials in six variables. However, this will not exclude the possibility that the corresponding real algebraic varieties may be equal. Hence we cannot exclude further exceptional cases in the realm $P = 0$ but $f \neq 0$ or $g \neq 0$. The isospectral classes in the case $f = g = 0$ will be one-dimensional: the constraint $f = g = 0$ allows one to express the variables S_{12}, S_{13}, S_{14} in terms of $I_3, D_{12}, D_{13}, D_{14}$. The remaining constraints $I_1 = i_1, I_4 = i_4$ define a family of curves obtained as intersections between spheres and tube-like surfaces.

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