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A discrete finite-dimensional phase space approach for the description of Fe8 magnetic clusters: Wigner and Husimi functions

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Abstract
A discrete quantum phase space formalism is used to discuss some basic aspects of the spin tunnelling which occurs in Fe8 magnetic clusters by means of Wigner functions as well as Husimi distributions. Those functions were obtained for sharp angle states and symmetric combinations of the lowest energy doublet—the particular tunnelling energy doublet—with the application of an external magnetic field. The time evolution of those functions carried out numerically allows one to extract valuable information about the dynamics of the states under consideration, in particular, the coherent oscillations associated with the particular energy doublet.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

The basic ideas governing the discrete phase space picture of quantum mechanics of physical systems described by finite-dimensional state spaces have been long established, and the concept of discrete Wigner functions and the discrete analogue of the Weyl–Wigner transformations are now recognized as important tools in dealing with those systems [1–3]. Within this description, the quasiprobability distribution functions are written in terms of a pair of discrete variables—for each degree of freedom—whose domains are finite sets of integer numbers which constitute a finite lattice that stands for a quantum discrete phase space. In this context, some attempts were also made in order to present a unified approach to the discrete quasiprobability distribution functions in the sense of obtaining the Glauber–Sudarshan, Wigner and Husimi functions as particular cases of s-parametrized discrete phase space functions [4–6]. In particular it was then shown that a hierarchical order among them through a smoothing process characterized by a discrete phase space function that closely resembles the role of the Gaussian function in the continuous phase space can be established [6]. From an operational point of view, it is clear that if we have a discrete Wigner function, the corresponding discrete Husimi distribution can also be directly obtained. Furthermore, discrete quasiprobability distribution functions in finite phase spaces have potential applications in quantum state tomography [6–8], quantum teleportation [6, 9–12], phase space representation of quantum computers [13], open quantum systems [14], quantum information theory [15] and quantum computation [16], where they are a natural tool to deal with the essential features of the inherent kinematics of the physical systems.

In the present paper we intend to show that the discrete phase space approach presented before [2, 5, 6, 17] can also be applied to the case of magnetic molecules, in particular to the Fe8 magnetic cluster [18], where certain important quantum phenomena, such as spin tunnelling [19], are currently considered because they are based on strong evidences coming from results of measurements with great resolution. That magnetic cluster has been deeply investigated in the last few years [20–22] and has been characterized as an useful tool in studying the importance of quantum effects in spin tunnelling. Among the interesting properties it presents, it must be remarked that it has a $J = 10\hbar$ spin associated with its magnetic moment, and that the anisotropy constants characterizing its structure are experimentally determined which allows one to construct a *bona fide* phenomenological Hamiltonian from which spectra and wavefunctions can be obtained. Besides, since it presents a well-determined crossover temperature below which the tunnelling of the magnetic moment is dominated by the quantum effects [21], the experimental data obtained in that temperature domain can give inestimable information about that quantum phenomenon. The application of external magnetic fields on the Fe8 cluster also gives additional information about its quantum behaviour and reveals interesting features of the energy spectra [23]. In this form, the quantum kinematical content as well as the proposed phenomenological Hamiltonian associated with this magnetic cluster provide the basic elements from which we can develop and apply the discrete phase space approach. As a consequence, we show that Wigner functions, as well as Husimi distributions, can be obtained which are described by a pair of complementary variables—connected by a discrete Fourier transform—namely, angle and angular momentum variables—so that they can therefore reveal quantum correlations between those quantum variables. These functions then allow us to recognize those correlations and, furthermore, to describe the role of those correlations on the behaviour of the tunnelling energy doublet and to study the effects of the application of external magnetic fields. It is also seen that the numerical procedure involved in this time evolution calculations does not introduce significant errors in the results when compared with those coming from the diagonalization of the Fe8 phenomenological Hamiltonian.

This paper is organized as follows. In section 2 we present a brief review of the discrete phase space approach, including the procedures through which we obtain the discrete Wigner functions as well as the Husimi distributions. The procedure of getting the time evolution of the Wigner function is also presented. The application of the formalism to the Fe8 magnetic cluster is carried out in section 3, and section 4 is devoted to the conclusions.

2. Discrete phase space formalism: a brief review

The possibility of describing nonclassical states and expectation values of quantum systems through a quantum phase space picture has been widely explored in the literature mainly in what concerns those physical systems described by continuous variables [24]. In this section we want to recall the main aspects of a possible quantum phase space picture especifically constructed for treating physical systems described by finite-dimensional state space.

2.1. Wigner functions and the Liouvillian

In the past, we have proposed a mapping scheme that allows us to represent any operator acting on a finite-dimensional state space as a function of integer arguments [2, 17] on a discrete N^2-dimensional phase space; in other words, we have extended the well-known Weyl–Wigner procedure [24] of operators mapping for finite N-dimensional Hilbert state space cases. In this form, we are then able to study the quantum properties of a given system of interest through the mapping of the relevant operators and studying the corresponding mapped functions. Of course we can also obtain discrete Wigner functions that represent state operators upon a discrete N^2-dimensional phase space [2]. In this connection, it is direct to see that we can also construct the Husimi distribution function, as well as the analogue of the Glauber–Sudarshan function if this is desired [5, 6].

From a general point of view, let us start by considering that for a complete description of a physical system described by a N-dimensional state space a set of N^2 operators is needed so as to permit a total construction of the dynamical quantities associated with that system. On the other hand, it must be recalled that, following Schwinger, it is possible to construct a set of N^2 operators that in fact constitute an operator basis in the respective operator space [25] associated with the physical system of interest. Therefore, we begin by recalling some of the main properties of the Schwinger formalism.

Let us consider an unitary operator $\mathbf{V}$ that acts cyclically on an orthonormal finite-dimensional set of basis state vectors as

$$\mathbf{V}|u_k\rangle = |u_{k-1}\rangle, \qquad k = 1, \ldots, N;$$

this operator satisfies, by its turn, the eigenvalue equation

$$\mathbf{V}|v_l\rangle = v_l|v_l\rangle,$$

where $v_l = \exp(2\pi \mathrm{i}\frac{l}{N})$, that is, the eigenvalues of V are the lth roots of the unity. Schwinger has also shown that another unitary operator $\mathbf{U}$ acts cyclically on the eigenvectors of V as

$$\mathbf{U}|v_l\rangle = |v_{l+1}\rangle, \qquad l = 1, \ldots, N.$$

Furthermore, similarly to the operator $\mathbf{V}$, this unitary operator $\mathbf{U}$ also satisfies an eigenvalue equation

$$\mathbf{U}|\tilde{u}_k\rangle = u_k|\tilde{u}_k\rangle,$$

where, again, $u_k = \exp\big(2\pi \mathrm{i}\frac{k}{N}\big)$. In fact, it was remarked [25] that this set of eigenvectors, $\{|\tilde{u}_k\rangle\}$, do in fact constitute the same set of states from which the construction begun, namely $\{|u_k\rangle\}$. Using these results it can be shown that the two orthonormal eigenvector sets are related by the Fourier coefficients

$$\langle u_k|v_l\rangle = N^{-\frac{1}{2}} \exp\left(2\pi \mathrm{i}\frac{kl}{N}\right).$$

The main point in this construction is that a set of N^2 operators can be obtained from these unitary operators through the products

$$\mathbf{S}(k,l) = \frac{\mathbf{U}^k\mathbf{V}^l}{\sqrt{N}} \exp\left(\mathrm{i}\pi\frac{kl}{N}\right), \qquad k,l = 0, \ldots, N-1, \tag{1}$$

which constitutes a basis in the operator space for the N-dimensional state space physical system. This is also known as the symmetrized Schwinger basis.

Now, the proposed approach elaborated in order to allow us to treat the family of physical systems described by finite-dimensional state spaces in a discrete quantum phase space is based on a mapping procedure that takes abstract operators and represents them as functions of integer variables in an analogous way the continuous Weyl–Wigner procedure does. In the present case, the discrete Weyl–Wigner transformation of a given operator **O** into its corresponding representative in the discrete quantum phase space is given by a trace operation [2, 17]

$$O(m,n) = \frac{1}{N} Tr[\mathbf{G}^{\dagger}(m,n)\mathbf{O}]. \tag{2}$$

In this expression $\mathbf{G}(m,n)$ is an operator basis which guarantees the mod N invariance in the discrete phase space, and is given by

$$\mathbf{G}(m,n) = \sum_{k,l=0}^{N-1} \frac{\mathbf{S}(k,l)}{\sqrt{N}} \exp\left[\mathrm{i}\pi\phi(k,l;N) - \frac{2\pi\mathrm{i}}{N}(mk+nl)\right], \tag{3}$$

where $\mathbf{S}(k,l)$ is the symmetrized, orthonormal and complete operator elements introduced by Schwinger. It is the phase

$$\phi(k,l;N) = N I_k^N I_l^N - k I_l^N - l I_k^N,$$

where I_r^N means the integral part of r with respect to N, that assures the mod N invariance of the operator basis. It is important to observe that this phase is irrelevant when we look for the mapping of a single operator, but it is of importance in describing, for instance, products of operators [26]. From a mathematical point of view, the labels of the sums must in fact run over a complete set of remainders so that, depending on the degree of freedom under consideration, the labels domains may also be symmetric, namely, $-\frac{N-1}{2} \leqslant k, l \leqslant \frac{N-1}{2}$.

It is also well known that the time evolution of a quantum system can be described by the von Neumann–Liouville equation for the density operator. Now, with the help of the expression for the mapped commutator of two operators, it is straightforward to obtain the mapping of that dynamical equation on the corresponding discrete quantum phase space equation. As already shown before [2, 17], the von Neumann–Liouville time evolution equation (we will consider hereafter $\hbar = 1$)

$$\mathrm{i}\frac{\partial}{\partial t}\boldsymbol{\rho} = [\mathbf{H}, \boldsymbol{\rho}]$$

is mapped onto

$$\mathrm{i}\frac{\partial}{\partial t}\mathcal{W}(u,v;t) = \sum_{r,s=-\ell}^{\ell} \mathcal{L}(u,v,r,s)\mathcal{W}(r,s;t), \tag{4}$$

where

$$\mathcal{L}(u,v,r,s) = 2\mathrm{i} \sum_{m,n,a,b,c,d=-\ell}^{\ell} \frac{h(m,n)}{N^4} \sin\left[\frac{\pi}{N}(bc-ad)\right] \mathrm{e}^{[\mathrm{i}\pi\Phi(a,b,c,d;N)]} \times \mathrm{e}^{\{\frac{2\pi\mathrm{i}}{N}[a(u-m)+b(v-n)+c(u-r)+d(v-s)]\}} \tag{5}$$

is identified with the discrete mapped expression of the Liouvillian of the system, $h(m,n)$ stands for the mapped expression of the here assumed time-independent Hamiltonian of interest, and the phase $\Phi(a,b,c,d;N)$ guarantees the mod N invariance. The function of integers $\mathcal{W}(r,s;t)$ is the discrete Wigner function which is obtained by mapping the density operator defined in the finite-dimensional state space; for instance, for a pure state

$$\boldsymbol{\rho}(t) = |\psi(t)\rangle\langle\psi(t)|$$

we have

$$\mathcal{W}(m,n;t) = \frac{1}{N} Tr[\mathbf{G}^{\dagger}(m,n)|\psi(t)\rangle\langle\psi(t)|].$$

With these assumptions and considering the time-evolved density operator to be written in the form of a series [27]

$$\rho(t) = \rho(t_0) - \mathrm{i}(t-t_0)[\mathbf{H},\rho(t_0)] + \mathrm{i}^2\frac{(t-t_0)^2}{2!}[\mathbf{H},[\mathbf{H},\rho(t_0)]] - \cdots, \tag{6}$$

we see that its mapped expression is given by

$$\mathcal{W}(u,v;t) = \sum_{r,s=-\ell}^{\ell} \mathcal{W}(r,s;t_0)\left\{\delta_{r,u}^{[N]}\delta_{s,v}^{[N]} + (-\mathrm{i})(t-t_0)\mathcal{L}(u,v,r,s)\right.$$
$$\left. + \frac{(-\mathrm{i})^2(t-t_0)^2}{2!}\sum_{x,y}\mathcal{L}(u,v,x,y)\mathcal{L}(x,y,r,s) + \cdots\right\}, \tag{7}$$

where now $\mathcal{W}(u,v;t)$ is the solution at time t of the von Neumann–Liouville equation for the Wigner function. In expression (7) those terms between curly brackets can be identified as the mapped propagator that associates the initial Wigner function at time t_0 with a time-evolved Wigner function at time t. We also see that the time evolution expression comprises sums of products of the arrays associated with the Liouvillian and the discrete Wigner functions taken over the discrete phase space. In other words, since $\mathcal{L}(u,v,r,s)$ is given, it is used for calculating the time propagation of the Wigner function as in series (7). Indeed, that expression is to be used iteratively, i.e., the Wigner function resulting from a propagation with a small Δt feeds the next step of the time propagation numerical process. Then, from a numerical point of view, to calculate the time evolution of a particular density operator we only have to construct the arrays associated with the Liouvillian and the Wigner function of interest at time t_0.

2.2. *The Husimi distribution function*

From another perspective, concerning the mapped expression of the density operator, we can also have at our disposal a discrete Husimi function. The formal way of obtaining this function in the present context was discussed in previous papers in which the continuous Cahil–Glauber formalism [28] was extended for finite-dimensional state spaces [5, 6]. Let us briefly review how to obtain the discrete Husimi distribution from a previously given discrete Wigner function. This can be accomplished by means of an s-parameterized mod (N) operator basis, which is given as

$$\mathbf{T}^{(s)}(m,n) = \sum_{\eta,\xi=-\ell}^{\ell} \mathrm{e}^{[\mathrm{i}\pi\phi(\eta,\xi;N) - \frac{2\pi\mathrm{i}}{N}(\eta m+\xi n)]}\frac{\mathbf{S}^{(s)}(\eta,\xi)}{\sqrt{N}}.$$

Here the new mod (N) invariant operator basis, $\mathbf{T}^{(s)}(m,n)$, is defined through a discrete Fourier transform of the extended operator basis, namely

$$\mathbf{S}^{(s)}(\eta,\xi) = [K(\eta,\xi)]^{-s}\mathbf{S}(\eta,\xi), \tag{8}$$

where $K(\eta,\xi)$ is a bell shaped function expressed as a product of Jacobi theta functions

$$K(\eta,\xi) = \{2[\vartheta_3(0|\mathrm{i}a)\vartheta_3(0|4\mathrm{i}a) + \vartheta_4(0|\mathrm{i}a)\vartheta_2(0|4\mathrm{i}a)]\}^{-1}\{\vartheta_3(\pi a\eta|\mathrm{i}a)\vartheta_3(\pi a\xi|\mathrm{i}a)$$
$$+ \vartheta_3(\pi a\eta|\mathrm{i}a)\vartheta_4(\pi a\xi|\mathrm{i}a)\mathrm{e}^{(\mathrm{i}\pi\eta)} + \vartheta_4(\pi a\eta|\mathrm{i}a)\vartheta_3(\pi a\xi|\mathrm{i}a)\mathrm{e}^{(\mathrm{i}\pi\xi)}$$
$$+ \vartheta_4(\pi a\eta|\mathrm{i}a)\vartheta_4(\pi a\xi|\mathrm{i}a)\,\mathrm{e}^{[\mathrm{i}\pi(\eta+\xi+N)]}\}$$

with $a = (2N)^{-1}$ and $\mathbf{S}(\eta, \xi)$ are the Schwinger basis elements already mentioned before. The complex parameter s that appears in the above expression is limited to the interval $|s| \leqslant 1$.

In analogy with the decomposition expression of an operator into an operator basis, what involves its representative defined on the discrete quantum phase space, we have now

$$\mathbf{O} = \frac{1}{N} \sum_{m,n=-\ell}^{\ell} O^{(-s)}(m,n)\mathbf{T}^{(s)}(m,n). \tag{9}$$

In particular, if we choose $\mathbf{O} = \boldsymbol{\rho}$ we see that for $s = 0$ this expression gives the mapping leading to the Wigner function, while for $s = -1$ it leads to the Husimi distribution.

In this formalism we also have the expression that allows us to obtain the Husimi distribution directly from the Wigner function, namely

$$\mathcal{H}(m,n) = \sum_{m',n'=-\ell}^{\ell} E(m'-m, n'-n)\mathcal{W}(m',n'),$$

where $E(m'-m, n'-n)$ characterizes the mapping kernel of the transformation, and it is given by

$$E(m'-m, n'-n) = Tr[\mathbf{T}^{(0)}(m,n)\mathbf{T}^{(-1)}(m',n')].$$

Due to this general character the mapping expression (9) that allows us to obtain the Husimi distribution, which is smoother than the Wigner function, must be also applied to obtain the mapped expressions of all the related quantum operators. It must be stressed that since the expectation values of the quantum operators we are interested in must be the same, independent of the degree of the smoothing process

$$\langle \mathbf{O} \rangle = \mathrm{Tr}[\mathcal{W}(m,n)\mathcal{O}_{\mathcal{W}}(m,n)] = \mathrm{Tr}[\mathcal{H}(m,n)\mathcal{O}_{\mathcal{H}}(m,n)],$$

the mapped expression of the associated operator involved in the average calculation is modified in such a way to account for possible discarded informations, related, for instance, to superposition or entanglement introduced by the smoothing process resulting in the Husimi distributions.

Regarding the time evolution, it is then clear that, once the Wigner function is given at any instant of time, the Husimi distribution can be obtained straightforwardly in the same way. Furthermore, regarding the system state, since the Husimi distribution has a well-behaved shape, we expect to obtain a clearer picture of the physical process of interest through its use.

3. Application: Fe8 magnetic cluster

The discovery of magnetic molecules, in particular the Fe8 magnetic cluster, which was first synthesized by Wieghardt *et al* [18], was a great contribution for the study of quantum effects in the dynamics of spin tunnelling [29]. Under proper temperature conditions, the pure quantum effects in the dynamics of the magnetization become manifest in some measurements performed with this molecule, specially due to the very well-measured constants characterizing its structural properties. The study of the orientation of the magnetic moment with these clusters also benefits from the fact that the presence of an external magnetic field may enhance some quantum transitions that allow one to test basic hypothesis about the quantum spin tunnelling.

The main experimentally determined results related to the Fe8 magnetic cluster indicate that it is a system with a $J = 10$ spin—that implies in a 21-dimensional state space for this degree of freedom—with an observed barrier of about 24 Kelvin (K). Furthermore, it was verified that below 0.35 K the relaxation of the magnetization becomes temperature independent [21], and that when an external magnetic field with intensity that is given by integer multiples of $\Delta H_{\parallel} \simeq 0.22$ Tesla (T) is applied along the easy-axis of the cluster (z-axis) the hysteresis curve has a behaviour [22] which can be interpreted as if there is an energy matching of states on both sides of a double-minima barrier, similar to what had already been proposed in another magnetic molecule, namely the Mn12ac cluster [30]. From the experimental data the proposed Hamiltonian for the Fe8 cluster (sometimes called giant spin model) in the presence of external magnetic fields has the form

$$\mathbf{H} = \mathbf{H}_0 + \mathbf{H}_1, \tag{10}$$

where the term

$$\mathbf{H}_0 = D\mathbf{J}_z^2 + \frac{E}{2}\left(\mathbf{J}_+^2 + \mathbf{J}_-^2\right)$$

constitutes the part that is established by the measured properties of the molecular structure, and the remaining ones

$$\mathbf{H}_1 = A\mathbf{J}_z + B(\mathbf{J}_+ + \mathbf{J}_-) + C(\mathbf{J}_+ - \mathbf{J}_-)$$

give the contribution that is associated with the external magnetic field. The anisotropy constants are [22] $D/k_B = -0.275$ K and $E/k_B = 0.046$ K, where k_B is the Boltzmann constant, while $A = g\mu_B H_{\parallel}$, $B = g\mu_B \cos\alpha H_{\perp}$ and $C = g\mu_B \sin\alpha H_{\perp}$ are the parameters associated with the magnetic field intensity along the z-axis (easy-axis), y-axis (medium-axis) and x-axis (hard-axis), respectively. Furthermore, this model Hamiltonian is expressed in terms of the angular momentum operators that obey the standard commutation relations

$$[\mathbf{J}_{\pm}, \mathbf{J}_z] = \mp\mathbf{J}_{\pm}, \qquad [\mathbf{J}_+, \mathbf{J}_-] = 2\mathbf{J}_z,$$

and its numerical diagonalization can be directly carried out within the 21 -dimensional set of $|j, m\rangle$ states, so that those energy eigenvalues thus obtained can be useful as reference values.

Now, the main requirement for the use of the proposed description of physical systems with finite-dimensional state space consists in characterizing the starting space state such that the quantum phase space mapping scheme is completely established. In the present case we have to establish a connection between our approach and the mathematical description of the proposed model for the Fe8 magnetic cluster. This connection, which is then the key element in order to allow the use of the quantum phase space description for the Fe8 cluster, is obtained just by choosing

$$|u_k\rangle \equiv |j, k\rangle$$

with the associated eigenvalue equation following the prescription proposed by Schwinger [25]

$$\mathbf{U}|j, k\rangle = \exp\left(2\pi \mathrm{i}\frac{k}{N}\right)|j, k\rangle, \tag{11}$$

where $\mathbf{U}$ is the unitary operator already discussed and $|u_k\rangle$ are its eigenstates. Furthermore,

$$\begin{aligned} \mathbf{J}_z|j, k\rangle &= k|j, k\rangle, \\ \mathbf{J}^2|j, k\rangle &= j(j+1)|j, k\rangle, \\ \mathbf{J}_{\pm}|j, k\rangle &= \sqrt{(j \mp k)(j \pm k + 1)}|j, k \pm 1\rangle, \end{aligned}$$

as usual, where now $-10 \leqslant k \leqslant 10$.

Therefore, we have a 21-dimensional space of the $\mathbf{U}$ eigenvectors as given above, and consequently we also have another 21-dimensional space, $\{|v_l\rangle\}$, of eigenvectors of $\mathbf{V}$, the complementary unitary operator also proposed by Schwinger. It is worth noting that the set of states $\{|v_l\rangle\}$, where $-10 \leqslant l \leqslant 10$, is in fact a basis since it is in this finite space that it is orthonormal and complete, thus it is also a *bona fide* basis. Since it was also shown [25] that the set of states $\{|v_l\rangle\}$ is obtained through a discrete Fourier transform of the basis states $\{|u_k\rangle\}$, the physical interpretation of the $\{|v_l\rangle\}$ states is then direct: they are associated with the polar angle orientation of the Fe8 cluster spin. Here the basis sets $\{|u_k\rangle\}$ and $\{|v_l\rangle\}$ are analogous to the canonical momentum and coordinate bases used in the continuous Wigner function.

Based on this association, we can now use the discrete Weyl–Wigner formalism discussed before to describe the quantum behaviour of the Fe8 cluster in a discrete 21^2-dimensional phase space. Thus far the kinematical content of the description has been already set up, then we can go one step further and obtain the mapped expression associated with the Fe8 cluster Hamiltonian (10). Following the mapping scheme presented before the discrete phase space expression for that Hamiltonian is

$$\begin{aligned} h(m,n) &= Am + Dm^2 + EQ(m,n)\cos\frac{4\pi n}{N} \\ &+ \sum_{k,r=-10}^{10} \frac{e^{\frac{2\pi i}{N}[r(m-k-\frac{1}{2})-n]}}{2N}\sqrt{(j-k)(j+k+1)}(B\cos\alpha + C\sin\alpha) \\ &+ \sum_{k,r=-10}^{10} \frac{e^{\frac{2\pi i}{N}[r(m-k+\frac{1}{2})-n]}}{2N}\sqrt{(j+k)(j-k+1)}(B\cos\alpha - C\sin\alpha), \end{aligned}$$

where

$$Q(m,n) = \sqrt{(j+m)(j-m+1)(j-m)(j+m+1)}.$$

With this result, it is straightforward now to find the expression for the Liouvillian. After a direct but tedious calculation, the discrete phase space expression for the Liouvillian is obtained, and it is written as

$$\begin{aligned} \mathcal{L}(u,v,r,s) = \frac{2i}{N^3}\sum_{m,a,c,d}\Bigg(\Bigg[&-(Am+Bm^2)\sin\left(\frac{\pi}{N}ad\right)e^{i\pi\Phi(a,0,c,d;N)} \\ &+C\sqrt{(j+m)(j-m+1)}\sin\left[\frac{\pi}{N}(c-ad)\right] \\ &\times e^{\frac{i\pi}{N}(a+2v)}\,e^{\frac{2i\pi}{N}c(r-a)}\,e^{i\pi\Phi(a,1,c,d;N)} \\ &-D\sqrt{(j-m)(j+m+1)}\sin\left[\frac{\pi}{N}(c+ad)\right] \\ &\times e^{\frac{-i\pi}{N}(a+2v)}\,e^{\frac{2i\pi}{N}c(r-a)}\,e^{i\pi\Phi(a,-1,c,d;N)} \\ &+\frac{Q(m,n)}{2}\left\{\sin\left[\frac{\pi}{N}(2c-ad)\right]e^{\frac{4\pi i}{N}v}\,e^{i\pi\Phi(a,2,c,d;N)}\right. \\ &\left.-\sin\left[\frac{\pi}{N}(2c+ad)\right]e^{\frac{-4\pi i}{N}v}\,e^{i\pi\Phi(a,-2,c,d;N)}\right\}\Bigg]e^{\frac{2\pi i}{N}[a(u-m)+c(u-r)+d(v-s)]}\Bigg). \end{aligned} \tag{12}$$

Having obtained the discrete Wigner function and using the mapped series, equation (7), we are then able to compute the temporal evolution of a state given at $\tau = 0$ with the expression above, equation (12).

In what concerns the Wigner function at $\tau = 0$, we may choose those that may be relevant for our purposes. At first, we start, for simplicity, from the density operator $\boldsymbol{\rho}$ for a pure state

$$\boldsymbol{\rho} = |\psi\rangle\langle\psi|,$$

where $|\psi\rangle$ can be expressed in terms of the eigenstates of the operator $\mathbf{U}$, equation (11), which are also, due to the adopted formalism, eigenstates of the angular momentum operators $\mathbf{J}^2$ and $\mathbf{J}_z$, namely

$$|\psi\rangle = \sum_{l=-10}^{10} C_l |j,l\rangle.$$

As has already been shown elsewhere [31], the Wigner function in this case is written as

$$\mathcal{W}(m,n) = \frac{1}{N} \sum_{r,s=-10}^{10} \exp\left[\frac{2\pi\mathrm{i}}{N}(mr+ns)\right] g(r,s), \tag{13}$$

where

$$g(r,s) = \frac{1}{N} \sum_{k=-10}^{10} C_k C^*_{\{k+s\}} \exp\left[-\frac{2\pi\mathrm{i}}{N} r\left(k+\frac{s}{2}\right)\right],$$

and the symbol $\{k+s\}$ stands for the sum cyclically restricted to the interval of labels, e.g., $k_{\max}+1 = k_{\min}$. As can be directly recognized, a great advantage of using the Wigner functions in this form is that, by diagonalizing the Hamiltonian in the angular momentum state basis $\{|j,k\rangle\}$, we can obtain the coefficients C_k to construct the Wigner function through equation (13).

3.1. Sharp angle state

A first illustrative case that can be directly treated is a sharp angle state associated with the physical configuration where, at $\tau = 0$, the magnetic moment of the Fe8 cluster points to a particular angle $\{\theta,\varphi\}$. The parallel and transverse magnetic fields are, in particular, $H_\parallel = 0.01T$, and $H_\perp = 2.0T$ respectively, being $H_\perp$ oriented at an angle of $\varphi = \pi/4$ relative to the x-axis, and the initial state, a sharp angle, is peaked at $n = 0$, where $\theta_n = \frac{\pi}{10}n$ with $-\pi \leqslant \theta_n \leqslant \pi$, and $-10 \leqslant n, m \leqslant 10$. The correspondent sequence of the time-evolved Wigner functions is depicted in figure 1.

Now, since we have the time-evolved Wigner functions for the peaked angle state, the associated Husimi distributions can be directly obtained, and are shown in figure 2.

In general, if the Wigner function evolves in time not clearly showing the behaviour of the physical system, the Husimi distribution, on the other hand, can afford for a better visualization of how the state evolves over the discrete phase space. In particular, it is to be noted the smeared behaviour that the Husimi distribution presents.

Also we see that, after a transient time—during which the initial distribution loses its identity—the peak of the distribution passes recurrently on the same regions of the $\theta = [0,\pi]$ sector of the discrete phase space.

It can be seen that since these functions are constructed on a discrete phase space—labelled by angle and angular momentum variables—they are also tailored to show the correlations between those variables, therefore complementing a description purely based on direct density matrix calculations involving only one quantum variable. The Wigner function in fact presents small negative values (of the order of 10^{-3}) which are direct manifestations of the presence of a symmetry in the Hamiltonian. Since $(-1)^{\mathbf{J}_z}$ commutes with the Hamiltonian, parity shows up in the angular momentum sector of the discrete phase space as the staggered behaviour. It is worth observing that not only superpositions or entaglement produce negative values in the Wigner function; discrete symmetries also can do this in a manifest way [17]. Furthermore, the Husimi distribution is obtained by a smearing process that involves only the next neighbour

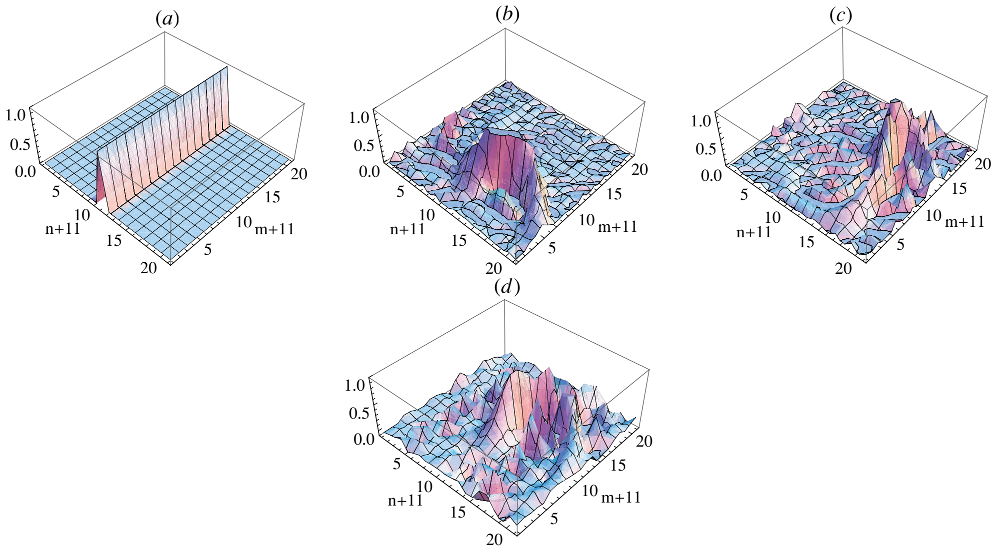

Figure 1. Time-evolved Wigner function calculated with time steps of $\Delta t = 0.05 \times 2\pi\mathrm{K}^{-1}$ and external magnetic fields $H_{\parallel} = 0.01$ T and $H_{\perp} = 2.0$ T oriented at an angle of $\phi = \pi/4$ with the x-axis. The labels m and n correspond to the finite-discrete angular momentum and angle variables respectively displaced 11 units of their original intervals. Figure (a) corresponds to the Wigner function at $\tau = 0$. Figures (b), (c), and (d) depict the Wigner functions for $\tau = 0.4 \times 2\pi\ \mathrm{K}^{-1}$, $0.8 \times 2\pi\ \mathrm{K}^{-1}$ and $1.2 \times 2\pi\ \mathrm{K}^{-1}$ respectively.

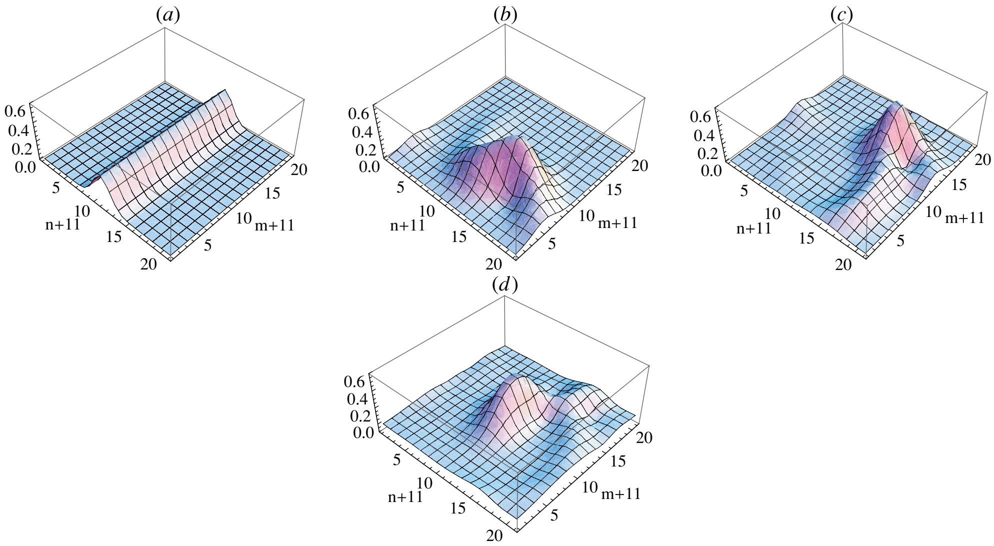

Figure 2. Time-evolved Husimi function under the same conditions indicated in figure 1.

site of the discrete phase space, and this is sufficient to wash out the staggering due to parity, but does not alter the main qualitative aspects of the Wigner function. Finally, the correlations between angle and angular momentum variables then manifest themselves in the dispersion of the Wigner and Husimi functions over the discrete phase space. By its turn, the particular external magnetic field used here confines those functions in specific regions of the discrete phase space.

3.2. Symmetric combination

As a second case we consider the symmetric combination, namely,

$$|\psi^s\rangle = \frac{1}{\sqrt{2}} \sum_k \left(C_k^i \left|u_k^i\right\rangle + C_k^j \left|u_k^j\right\rangle\right),$$

where the labels i and j characterize the Fe8 cluster energy eigenstates of which the combination is made of. Here we will be interested in the energy doublet constituted of the ground state and its neighbour partner. This choice is based on the fact that this doublet is the proposed candidate for showing the spin tunnelling properties when the Fe8 cluster is submitted to temperatures below the well-established crossover temperature. Furthermore, it is also known that the energy gap between them is widened when an external magnetic field—parallel to the easy-axis—is applied on the cluster. In particular, this study is of great interest when this external magnetic field, $H_\parallel$, is an integer multiple of 0.22 T because of the matching of energy levels as pointed out before.

For those states it is also simple to obtain the Wigner function by means of equation (13), namely $\mathcal{W}^s(m, n)$, and propagate it in time. In figure 3 we depict a sequence of time-evolved Wigner functions for $H_\parallel = 0.11$ T introduced in order to induce a convenient widening of the energy gap, and $H_\perp = 0.0$ T. For this situation we have, through the diagonalization of the Hamiltonian, the reference values $E_g = -29.017\,45$ K and $E_1 = -26.064\,41$ K which are the ground and the first excited states respectively. The energy gap between those states is then $\Delta E_{\text{ref}} = 2.953\,04$ K.

First of all we observe that the staggered behaviour of the Wigner function, mainly visible in the central region of the discrete phase space, is again due to parity effects related to the angular momentum variable. Moreover, what is important here is that the initial configuration is totally recovered after $\tau \approx 2.15 \times 2\pi$ K^{-1}, as can be promptly recognized; this periodicity with time of the Wigner function indicates that we have a coherent oscillation that is associated with the magnetic moment quantum tunnelling.

If, instead of the Wigner function, we have the family of the corresponding Husimi distributions, as shown in figure 4, we immediately see that the staggering was smeared. Also, as an intended result, the oscillation can be clearly seen to occur along the angle sector of the phase space such that the periodicity is obtained and agrees with that observed with the Wigner function; in other words, the smearing process given rise to the Husimi distributions intentionally preserved the main aspects contained in the Wigner function.

Note that, the periodicity of the Wigner function associated with that combination of states could also be obtained from the time correlation function defined as

$$P_{if}(\tau) = \sum_{m,n} \mathcal{W}^i(m, n; 0)\mathcal{W}^f(m, n; \tau).$$

A calculation performed with the same external magnetic field as before ($H_\parallel = 0.11T$ and $H_\perp = 0.0T$) gives $P_{if}(\tau)$ as shown in figure 5. Once again we obtain for the symmetric state a periodicity in time of $\tau \approx 2.15 \times 2\pi \text{K}^{-1}$. Of course the oscillation period can be obtained from figure 4 as well from figure 5; both coincide.

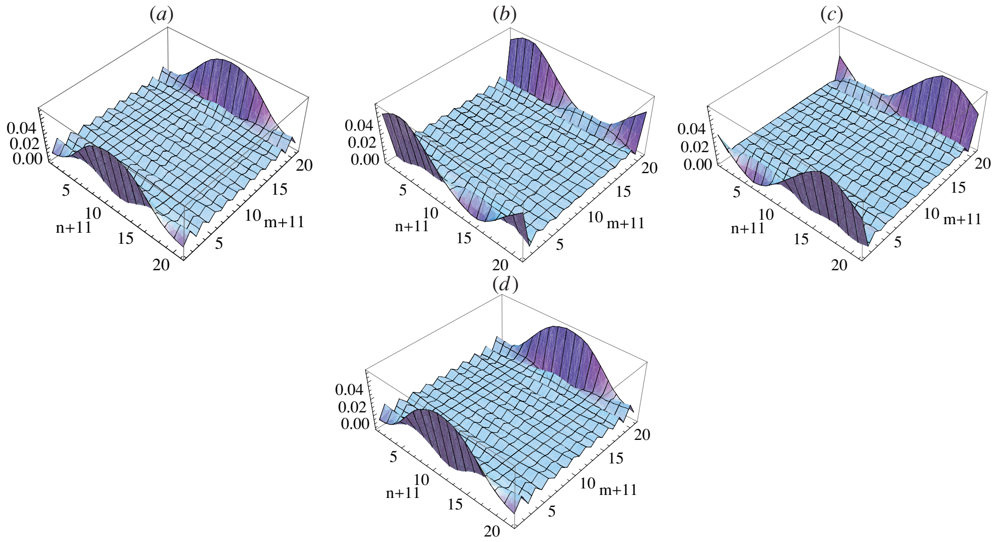

Figure 3. Time evolution of the Wigner function generated by a symmetric combination of the lowest energy levels. The time step was taken as $\Delta t = 0.05 \times 2\pi$ K^{-1}, and the external magnetic fields intensities were $H_\parallel = 0.11$ T and $H_\perp = 0.0$ T. Here the labels m and n also are the discrete angular momenta and angle variables. Figure (a) corresponds to the Wigner function at $\tau = 0$. Figures (b), (c) and (d) depict the Wigner functions for $\tau = 0.75 \times 2\pi$ K^{-1}, $1.55 \times 2\pi$ K^{-1} and $2.15 \times 2\pi$ K^{-1}, respectively.

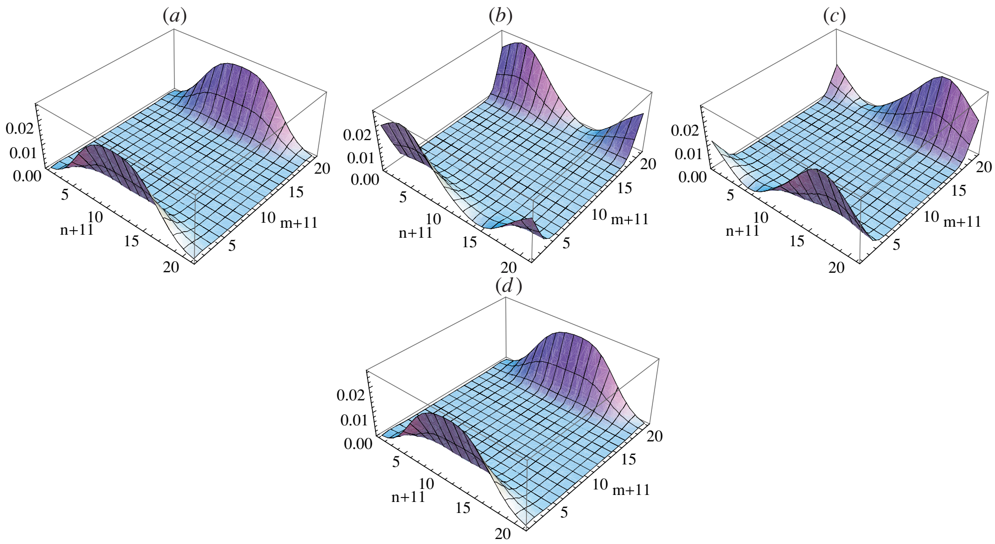

Figure 4. Time-evolved Husimi distribution under the same conditions indicated in figure 3.

Now, as an interesting result related to this we see that we can associate this oscillation period, $\tau(H_\parallel)$, with the energy gap between the ground state and its neighbour partner—which give the first energy doublet—by just using

$$\Delta E(H_\parallel) = \frac{2\pi}{\tau(H_\parallel)}, \tag{14}$$

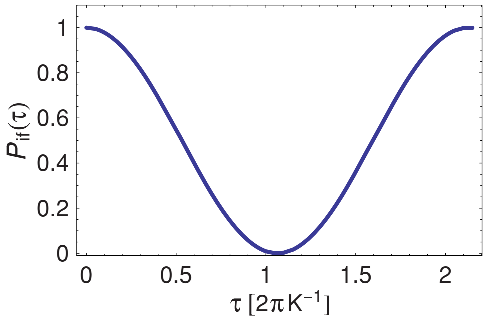

Figure 5. Time correlation function calculated with time step of $\Delta t = 0.05 \times 2\pi$ K^{-1}. The external magnetic field intensities are $H_{\|} = 0.11$ T and $H_{\perp} = 0.0$ T.

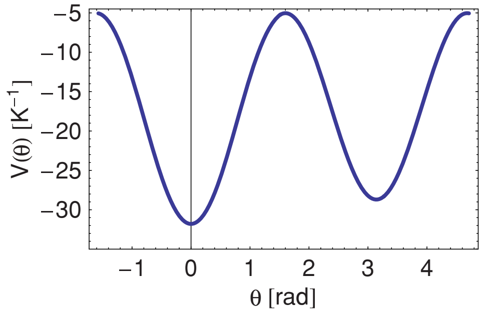

Figure 6. Potential function of the Fe8 cluster obtained through an angle-based approach. The magnetic field intensities are $H_{\|} = 0.11$ T and $H_{\perp} = 0.0$ T.

where we can associate a frequency $\omega(H_{\|})$ with the oscillation period, and $\Delta E(H_{\|})$ is the energy gap given as a function of the magnetic field intensity. It is important to emphasize that this relation is dependent on the magnetic field intensity, and that here we want to consider only weak magnetic field intensities effects on the tunnelling spin energy doublet. For the values of the above considered external magnetic field and using $\tau = 2.15 \times 2\pi$ K^{-1}, we obtain from equation (14) $\Delta E = 2.922\,41$ K, while the reference value is $\Delta E_{\mathrm{ref}} = 2.953\,04$ K, thus showing a deviation of 1.04%, which indicates a good agreement.

Still along the same line we have just followed, and in order to further clarify the tunnelling process occurring in this case, we can also use an angle-based approach developed in recent years [32, 33] to describe this same physical situation. In that approach we obtain an effective Hamiltonian, written in terms of a potential and an effective mass function, that can describe the present situation and that can also complement the present discussion. To highlight the main spin tunnelling characteristics visualized in figures 4 and 5 we plot the associated potential function, figure 6,

$$V(\theta) = (D + E)J(J + 1)\cos^2(\theta) - g\mu_B H_{||}\sqrt{J(J + 1)}\cos(\theta) - EJ(J + 1),$$

where the Fe8 parameters are those already presented before. We then see that the peaks of the wavefunctions which generate the Husimi distribution—associated with the symmetric combination at $\tau = 0$—occur at both deep minima at $\theta = 0$ and $\theta = \pi$ of the potential. It is then immediate to verify that the leakage of the Husimi distribution into angular regions that would not be classically accessible indicates that tunnelling is occurring.

4. Conclusions

Based on a formal approach aiming at the construction of a discrete phase space picture of quantum mechanics related to finite-dimensional state spaces developed before, and briefly reviewed here, we addressed the problem of spin tunnelling in the Fe8 magnetic cluster. Our formal approach was shown to hold also in this particular case of finite-dimensional quantum system, and starting from the phenomenological Hamiltonian describing the magnetic cluster with or without an external magnetic field we obtained the discrete phase space representatives of the relevant elements which govern the complete description of the system and its time evolution. The Liouvillian associated with the Hamiltonian—which propagates the density operator in time in the discrete phase space—was explicitly written. In what concerns the representation of the state of the Fe8 magnetic cluster we have shown how a discrete Wigner function, as well as a discrete Husimi distribution, can be directly obtained.

The time evolution of those functions for the Fe8 magnetic cluster was studied in two different cases: (a) a sharp angle state with an external magnetic field and (b) symmetric combination of two energy eigenstates of the phenomenological Hamiltonian; in particular, we chose the lowest energy doublet. In both cases the Wigner function, as well as the Husimi distribution, were calculated and relevant information concerning the dynamics of the spin was extracted from them. With respect to the time evolution of the symmetric combination of states, it was also shown that, by analysing the periodicity of its oscillatory behaviour, a period could be found that allowed us to calculate the energy gap associated with the tunnelling doublet of states from which we started.

The results presented here corroborate all the basic formal scheme proposed before as well as the procedures adopted here to describe spin systems, in particular the Fe8 magnetic cluster, in the corresponding discrete phase space.

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