Entanglement properties in molecules and low-dimensional magnetic systems: general theoretical guidelines and some specific proposals for the experimental analysis based on Neutron and Muon Scattering

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(Dated: July 7, 2010)

During the period 10 May - 7 June 2010 I've been working at the ISIS Neutron Pulse Source of the Rutherford Appleton Laboratory (Didcot, Oxford, UK). A report of the research activity, focused on the possibility of experimentally observe features related with entanglement properties in solid-state and molecular systems, follows. In particular, in Section I we describe our proposal for a Neutron Scattering experiment for observing the phenomenon of complete entanglement suppression via an external magnetic field. In Section II we present an original theoretical approach that allows us to define some general guidelines for the experimental observation of entanglement properties.

I. ENTANGLEMENT-SWITCH: A PROPOSAL FOR EXPERIMENTAL ANALYSIS VIA NEUTRON SCATTERING

This part of the activity has been developed in collaboration with the research group of Prof. T.Perring at the ISIS Neutron Pulse Source of the Rutherford Appleton Laboratory, and also discussed with Prof. A. Boothroyd of the Physics Department of the Oxford University. Most of the work has been focused upon the possibility of submitting a proposal for a Neutron Scattering experiment to be done at the ISIS Neutron Source. To this respect, necessary requisites and preferred conditions are marked differently (bold and italic, respectively) in what follows.

A. Setup

This is the essential scheme as far as the real compound to be tested and the necessary laboratory facilities are concerned.

- A single chrystal whose magnetic behaviour can be effectively described as a spin system on a d-dimensional **bipartite** lattice of N^d sites;
- On each site i sits a spin $\boldsymbol{S}_i$, which **interacts only with its z nearest neighbours via a Heisenberg-like exchange interaction**

$$\mathcal{H}_0 = \sum_{<ij>} J_x S_i^x S_j^x + J_y S_i^y S_j^y + J_z S_i^z S_j^z \ , \qquad (1)$$

(the sum is over all pairs of nearest neighbours).

- A uniform magnetic field $\boldsymbol{h} = (h_x, h_y, h_z)$ is applied along a given **known direction**, inducing a Zeeman term interaction

$$\mathcal{H}_{Ze} = \sum_i \boldsymbol{h} \cdot \boldsymbol{s}_i \qquad (2)$$

(all constants are set equal to unity).

- The neutron scattering cross section is measured in specific directions, which must be determined relatively to those of the chrystallografic axis and that of the applied field.

B. Theoretical statement

It is rigorously demonstrated[1] that for a magnetic system with Hamiltonian $\mathcal{H}_\prime + \mathcal{H}_{\rm Ze}$, if

$$\sum_\alpha \frac{h_\alpha^2}{(J_\alpha + J_\beta)(J_x + J_\gamma)} = zS \ , \qquad (3)$$

where greek indeces get values x, y, z with $\beta \neq \alpha$, $\gamma \neq \alpha$ and $\beta \neq \gamma$, then it is

$$|GS\rangle = \Pi |s_i\rangle \qquad (4)$$

where $|GS\rangle$ is the ground-state of the system, and $|s_i\rangle$ are single-spin pure states, which are eigenstates of $\boldsymbol{n}_i \cdot \boldsymbol{S}_i$, where the local spin-orientation $\boldsymbol{n}_i$ is determined by the Hamiltonian parameters J_α and $\boldsymbol{h}$. In particular, if S=1/2 it is

$$|s_i\rangle = \cos\theta_i e^{-i\varphi_i} |\uparrow\rangle + \sin\theta_i e^{i\varphi_i} |\downarrow\rangle \qquad (5)$$

where θ_i and φ_i are fully determined by J_α and $\boldsymbol{h}$. When the exchange integrals are uniform along the chain (as in Eq. (1)) the dependence upon the index i reduces to a dependence on whether the site belongs to one or the other of the two sublattices (remember that the lattice MUST be bipartite), so that only two directions, $\boldsymbol{n}_{1,2}$, are defined. The field defined by Eq. (3) is usually called "factorizing" field ($\boldsymbol{h}_f$); without loss of generality, we will hereafter set its direction to be either x or z so as to drop the vector notation whenever possible.

The above result holds in the *antiferromagnetic case* (preferred) ($J_x > 0, J_y > 0, J_z > 0$); however, a generalization to the ferromagnetic case is also available. The value of the spin is not particularly relevant, but *low values of S* are to be preferred ($S = 1/2$ or 1).

C. Consequences on Entanglement properties

The ground state of the system at h_f is fully separable and therefore is, by definition, a non entangled many-body ground-state (sometimes referred to as a "classical-like" ground state). When the structure of $|GS\rangle$ is not trivial (as it is, for instance, in the case of the fully aligned ferromagnetic ground state), the above result stands as a very striking feature for a strongly interacting quantum system.

In fact, despite standard magnetic observables (such as correlation functions and magnetizations) do not show peculiar behaviour at or near h_f, entanglement properties do: not only all the entanglement measures vanish when $h = h_f$, but they show a very peculiar field dependence, characterized by cusp-like behaviour, both in one [2] and two dimensions [3].

This result did and still does resonate a lot in the scientific community, both from the point of view of entanglement and from that of magnetic models and systems. However, no experimental analysis of the overall phenomenon has been proposed yet, possibly due to the fact that ground-state properties are only accessible at $T = 0$, but also due to the lack of direct consequences of the very peculiar structure of the ground state upon some measurable quantity.

D. Consequences on $S(q,\omega)$ @ $h = h_f$ (exact)

In fact, as demonstrated by Mueller and Shrock in 1985 [4], *"the special structure of the ground-state wavefunction at $h = h_f$ has the consequence that the three diagonal dynamic structure factors $S_{\mu\mu}(q,\omega)_H$, $\mu = x, y, z$ are expressible in terms of a single function $S_{+-}(q,\omega)_{\widetilde{H}}$, where $\widetilde{H}$ is the Hamiltonian of the system with the quantization axis on site i chosen parallel* to $\boldsymbol{n}_i$ (see Sec.II of Ref. 4 for details). Would experimental data for $S_{\mu\mu}(q,\omega)_H$ at h_f be available one could first check the above result, even without knowing the function $S_{+-}(q,\omega)_{\widetilde{H}}$. Moreover, the function $S_{+-}(q,\omega)_{\widetilde{H}}$ is analitically accessible in some specific case, namely

i) for any q in the XXZ model ($J_x = J_y$);

ii) for $q = 0$ in the generic XYZ model.

In the above two special cases one could plot experimental data together with theoretical ones from Eqs.(3.8a-c) of Ref. 4 and check the overall picture.

Problem with this approach is that despite being exact, the results presented in Ref. 4 do only hold for $h = h_f$ and $T = 0$: discrepancies with respect to the expected picture could hence arise from this condition not being exactly fulfilled, a possibility which could make the overall analysis tricky and unstable.

E. Consequences on $S(q,\omega)$ @ $h \simeq h_f$ (modified spin-wave theory)

Our proposal is based on the results obtained by a modified spin-wave theory (SWT), to be possibly refined up to some self-consistent level, where the quantization axis at site i is chosen to be parallel to the alignment direction defined by $\boldsymbol{n}_i$. For $h \simeq h_f$ this comes as a natural assumption, given the structure of $|GS\rangle$ at h_f. The resulting modified spin wave theory, despite being slightly more complicated than the usual one, produces readable expressions. Depending on the model chosen, some pathologies of the theory arise, in particular for $d = 1$. On the other hand, there are "healthy" situations, amongst which we particularly mention

1) any two dimensional antiferromagnet, with field applied in any directions.

2) one dimensional anisotropic antiferromagnet, with field applied perpendicular to the direction of the anisotropy (as in the case, for instance, of the XXZ model with field applied along the X direction, $J_x < J_x, J_y$ and $\boldsymbol{h} = (h_x, 0, 0)$).

In fact, models with a gap (Δ) in the excitations spectrum are to be preferred, as this (besides granting stability to the harmonic approximation underlying the SWT) gives a more precise meaning to the condition $T = 0$, which in fact becomes $T < \Delta$.

The proposed modified SWT not only works at $h = h_f$ but it is a reasonable approximation also for h in the vicinity of h_f: therefore, results for different values of the field becomes available and an analysis of what in fact happens at $h = h_f$ gets possible. In particular, we have shown that the component of the dynamical structure factor $S_{z'z'}(q,\omega)$ along the quantization axis defined by $\boldsymbol{n}_1$ (hereafter named z') exactly vanishes for all k and ω when $h = h_f$.

Analytical, despite approximated, expressions for the components of the dynamical structure factors in a finite interval of h-value centered at h_f are available via the modified SWT: an experimental analysis for different values of h could hence be theoretically interpreted, possibly leading to the first experimental observation of an "entanglement-switch" phenomenon.

F. Experimental guidelines

The experiment should be based on both elastic and inhelastic neutron scattering on a single chrystal whose magnetic behaviour is known to be properly described by a Heisenberg, possibly anisotropic, Hamiltonian. A uniform magnetic field should then be applied, with value ranging in a small interval centered at h_f. Angles between the direction of the applied field and the characteristic magnetic axis of the chrystal must be precisely determined in order to define the laboratory framework for the neutron scattering (i.e. the direction $\boldsymbol{n}$ defined above). The dynamical structure factors along $\boldsymbol{n}$ and

the other two perpendicular directions should then be measured. The resulting data, once reported versus ω and q, together with the theoretical predictions, should confirm the predicted vanishing of $S(q,\omega)$ along $\boldsymbol{n}$ at h_f for whatever ω and q. The experiment should be done at low enough temperatures so as to grant both the low-dimensional character of the magnetic behaviour and the possibility to observe features which are essentially related with the ground state structure.

G. Possible compounds

- one-dimensional S=1/2: Cs_2CoCl_4 [8]
- one-dimensional S=1: anisotropic heisenberg antiferromagnets, possibly with single ion anisotropy, such as NENP [9] or NDMAP [10]
- two-dimensional S=1/2: monolayer copper oxides (such as La_2CuO_4 and $Sr_2CuO_2Cl_2$ and many others); $[Cu(pyz)_2(HF_2)]PF_6$ [13].

II. ENTANGLEMENT PROPERTIES: GENERAL THEORETICAL GUIDELINES FOR EXPERIMENTAL OBSERVATION

This part of the activity has been developed in collaboration with the research group of Dr. N.Gidopoulos at the ISIS Neutron Pulse Source of the Rutherford Appleton Laboratory. We have worked on a theoretical approach for describing open quantum systems, which might shed light upon the meaning of any parametric representation, as well as on the relation between the emergence of a Berry phase and Entanglement properties. In particular, aimed at determining quantum hermitian operators corresponding to physical observables possibly related with Entanglement, we have developed the following formalism.

Let us consider a composite system $S = AuB$ in a pure state $|\Psi\rangle$. We introduce an orthonormal basis $\{|\phi_\beta\rangle\}$ for the subsystem B and write

$$|\Psi\rangle = \sum_\beta \gamma_\beta |\phi_\beta\rangle \hat{O}_A^{\psi\beta} |\chi_A\rangle = \tag{6}$$

$$= |\Phi_B^{(A)}\rangle |\chi_A\rangle \ , \tag{7}$$

where $|\chi_A\rangle$ is any pure state of subsystem A, γ_β are complex coefficients and by the suffix (A) in $|\Phi_B^{(A)}\rangle$ we understand that despite $|\Phi_B^{(A)}\rangle$ formally representing a pure state for the subsystem B, it in fact depends parametrically on the state of the subsystem A. Such *parametric* dependence is embodied in the operators $\hat{O}_A^{\Psi\beta}$ which locally act on the subsystem A only (for the sake of a lighter notation, we will hereafter drop the index A). A matrix representation of the operators $\hat{O}^{\Psi\beta}$ is directly obtained by choosing an orthonormal basis for the subsystem A, say $\{|\chi_\alpha\rangle\}$, so that one can write

$$|\Psi\rangle = \sum_{\alpha\beta} c_{\alpha\beta} |\chi_\alpha\rangle |\phi_\beta\rangle \ , \tag{8}$$

and get the relations

$$\sum_{\alpha'} O^{\Psi\beta}_{\alpha\alpha'} \chi_{\alpha'} = \frac{c_{\alpha\beta}}{\gamma_\beta} \ , \tag{9}$$

for the matrix elements $O^{\Psi\beta}_{\alpha\alpha'} \equiv \langle\alpha'|\hat{O}^{\Psi\beta}|\alpha\rangle$. Normalization of $|\Phi_B^{(A)}\rangle$ implies

$$\sum_\beta |\gamma_\beta|^2 \ (\hat{O}^{\Psi\beta})^\dagger \hat{O}^{\Psi\beta} = \hat{\mathbb{I}} \ , \tag{10}$$

which clearly does not necessarily mean unitarity of each $\hat{O}^{\Psi\beta}$. If the Schmidt basis are used, so as to write $|\Psi\rangle = \sum_j v_j |\nu_j^A\rangle |\nu_j^B\rangle$, it is

$$\hat{O}^{\Psi j} = v_j \hat{\mathbb{I}} \quad \text{and} \quad (\hat{O}^{\Psi j})^\dagger \hat{O}^{\Psi j} = |v_j|^2 \hat{\mathbb{I}} \ , \tag{11}$$

from which it is easily seen that if $|\Psi\rangle$ is not entangled, i.e. if $v_j = \delta_{jl}$, then it is $\hat{O}^{\Psi j} = \delta_{jl}\hat{\mathbb{I}}$, and the whole construction is consistently trivial. On the other hand, if $|\Psi\rangle$ is maximally entangled, i.e. if $v_j = 1/\sqrt{N_A}$ for all j (here N_A is the dimension of the Hilbert space of subsystem A), then all the $\hat{O}^{\Psi j}$ are equal, and proportional to I.

Given any physical observable $\mathcal{M}$, and the corresponding Hermitian operator $\hat{M}$, the above formalism allows one to write

$$\langle\Psi|\hat{M}|\Psi\rangle = \langle\chi_A|\hat{M}_{\text{eff}}|\chi_A\rangle \ , \tag{12}$$

where we have introduced the "effectively local" operator $\hat{M}_{\text{eff}}$, defined by

$$\hat{M}_{\text{eff}} = \sum_{\beta'\beta} (\hat{O}^{\Psi\beta'})^\dagger \left(\gamma^*_{\beta'}\gamma_\beta \langle\phi_{\beta'}|\hat{M}|\phi_\beta\rangle\right) \hat{O}^{\Psi\beta} \ . \tag{13}$$

The reason why we call these operators "effectively local" is that despite their acting solely on the subsystem A, they do also depend on the global state $|\Psi\rangle$. We underline that the hermitianicity of $\hat{M}$ implies that of $\hat{M}_{\text{eff}}$, due to condition (10), i.e. due to the normalization of $|\Phi_B^{(A)}\rangle$. We now apply the above formalism to a well known example of entangled state.

Let us consider the case when A and B are qubits, i.e. physical objects that can be described as $S = 1/2$ spins, and chose $\{|\phi_\beta\rangle\} = \{|\chi_\alpha\rangle\} = \{|1\rangle, |0\rangle\}$. The state of the qubits pair be

$$|\Psi\rangle = x|10\rangle + y|01\rangle \tag{14}$$

where x and y are complex coefficients such that $|x|^2 + |y|^2 = 1$ (we will hereafter set x real). The two operators

$\hat{O}^{\Psi 1}$ and $\hat{O}^{\Psi 0}$ can be determined by Eq. (9). In matrix representation the result is

$$\hat{O}^{\Psi 1} \to \begin{pmatrix} |\chi_0| e^{i\varphi^1} & -\frac{\chi_1 |\chi_0|}{\chi_0} e^{i\varphi^1} \\ y\frac{\chi_1^*}{\gamma_1} & y\frac{\chi_0^*}{\gamma_1} \end{pmatrix} \tag{15}$$

and

$$\hat{O}^{\Psi 0} \to \begin{pmatrix} x\frac{\chi_1^*}{\gamma_0} & x\frac{\chi_0^*}{\gamma_0} \\ -\frac{\chi_0 |\chi_1|}{\chi_1} e^{i\varphi^0} & |\chi_1| e^{i\varphi^0} \end{pmatrix} \tag{16}$$

The phasis φ^1 and φ^0 entering the above expressions are undetermined and essentially arbitrary. We can therefore equal φ_1 to the phase of χ_0, i.e. $\exp(i\varphi_i) = \chi_0/|\chi_0|$, and φ_0 to that of χ_1, i.e. $\exp(i\varphi_0) = \chi_1/|\chi_1|$, thus getting

$$\hat{O}^{\Psi 1} \to \begin{pmatrix} \chi_0 & -\chi_1 \\ y\frac{\chi_1^*}{\gamma_1} & y\frac{\chi_0^*}{\gamma_1} \end{pmatrix} \quad \text{and} \quad \hat{O}^{\Psi 0} \to \begin{pmatrix} x\frac{\chi_1^*}{\gamma_0} & x\frac{\chi_0^*}{\gamma_0} \\ -\chi_0 & \chi_1 \end{pmatrix} \tag{17}$$

From Eqs. (13) and (17) we can derive the effectively local operator corresponding to any operator acting on both qubits. Notice that when $\hat{M}$ is a local operator, i.e. it only acts on one of the two subsystems, then it consistently is $\hat{M}_{\text{eff}} = \hat{M}$. In order to clarify the meaning of the effectively local operators, we have specifically addressed the case when $\hat{M}$ is the hamiltonian $\hat{\mathcal{H}}$ ruling the qubits interaction, that we have chosen of the Heisenberg form (for the sake of simplicity, the symbol over letters indicating the operatorial character of the object, will be hereafter implied).

$$\mathcal{H} = \boldsymbol{\sigma}_A \cdot \boldsymbol{\sigma}_B = 2(\sigma_A^+ \sigma_B^- + \sigma_A^- \sigma_B^+) + \sigma_A^z \sigma_Z^z \ . \tag{18}$$

After some algebra, we find

$$\mathcal{H}_{\text{eff}} \to \begin{pmatrix} |\chi_0|^2(q-p) + p & \chi_0^* \chi_1 (p-1) \\ \chi_0 \chi_1^* (p-1) & -|\chi_0|^2(q-p) + q \ , \end{pmatrix} \tag{19}$$

where $q = |x\gamma_0|^2 + |y\gamma_1|^2$ and $p = 2(xy^* + x^*y) - 1$. The dependence of $\mathcal{H}_{\text{eff}}$ on the state of the subsystem which it acts on (i.e. on the coefficients $\chi_{0,1}$) reflects the self-consistent character of the proposed formalism. As a consequence of such dependence, it is also $\mathcal{H}_{\text{eff}}|\chi_A\rangle = \widetilde{\mathcal{H}}_{\text{eff}}|\chi_A\rangle$, with

$$\widetilde{\mathcal{H}}_{\text{eff}} = (q-1)|\chi_1|^2 \sigma^z + (p+q-1)\hat{\mathbb{I}} \ . \tag{20}$$

The above expression shed some light on the actual meaning of effectively local operators: they stand as a formal tool for describing the effect that the very same existence of one subsystem has on the other, due to their global state being entangled. In the example above, such effect is represented by qubit A being subjected to an effective magnetic field along the z direction, due to its being interacting with qubit B. Notice that, if Ψ were NOT entangled (for istance if $y = 0$), than the effective field would vanish, and $\mathcal{H}_{\text{eff}}$ would reduce to $-\hat{\mathbb{I}}$ (and $\langle\Psi||\mathcal{H}||\Psi\rangle = -1$, as due), confirming that the non triviality of the effectively local operators is related with the state of the global system being entangled rather than on the two subsystems being interacting. We consider the above formalism a solid basis for developing an original analysis of parametric representation of open quantum systems, which might pave the way to a deeper understanding of the connection between geometric phases and entanglement. The work is in progress.

III. APPENDIX

During the period this report refers to I have also had fruitful discussions with Dr. Nicola Tartoni of the Diamond Light Source (Chilton, Oxford) and scientists in the research group of Prof. V.Vedral at the Clarendon Laboratory of the Department of Physics of the University of Oxford. I have attended several group meetings and seminars both at ISIS and at the Clarendon Laboratory. Moreover, I finalized and submitted a paper I'd been working on in the last six months in Florence [14] and arranged to give seminars at the Physics Department of the University College London (8th of July), the Department of Materials of the Oxford University (14th of July) and finally at ISIS (20th of July).

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