Chirality of Triangular Antiferromagnetic Clusters as a Qubit

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We show that the chirality of triangular antiferromagnetic clusters can be used as a qubit even if it is entirely decoupled from the total spin of the cluster. In particular, we estimate the orbital moment associated with the chirality, and we show that it can be large enough to allow a direct measurement of the chirality with a field perpendicular to the cluster. Consequences for molecular magnets are discussed, and an alternative implementation with Cu atoms on a surface is proposed, for which one- and two-qubit gates are worked out in detail. Decoherence effects are also discussed.

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It is widely recognized that the use of quantum properties of matter to store and treat information could bring exciting new possibilities (see, e.g., [1] and references therein). It is therefore important to find simple quantum systems (such as qubits, two-level systems) on which some essential basic operations could be made reliably. Requirements include the possibility to prepare and measure the system in a specific basis, and also to couple subsystems to perform coherent transformations of at least two qubits. At the same time, it should be free enough from decoherence to allow significant numbers of local operations to be performed, and should also be scalable to allow for large arrays of qubits. These requirements are very hard to fulfill, and many systems lack at least one of them. Solid state implementations have the potentiality to be scalable to large size, and have been actively studied, using, e.g., Josephson junctions [2], electronic spin in quantum dots [3,4], nuclear spin chains [5], or single-molecule magnets [6].

In that respect, antiferromagnetic triangular clusters are particularly interesting. First of all, a number of experimental realizations are already available. But more importantly, the presence of an extra degree of freedom in addition to the total spin, the chirality, opens new possibilities with respect to purely magnetic qubits. A first realization has been explored recently in [7]. There it was shown that the chirality induces a spin-electric effect whereby the total spin of the cluster can be rotated by an external electric field when spin-orbit interaction is taken into account. In that scheme, the qubit is still essentially magnetic, and the measurement relies on the spin magnetic moment of the qubit.

In this Letter, we discuss the alternative possibility of qubits entirely based on the chirality. Getting rid of the spin is of course a major advantage regarding decoherence since such a qubit would be less sensitive to magnetic noise (see below), but this creates at the same time a potential problem for measurement since there is no Zeeman coupling of the qubit to an external magnetic field any more. As we show, the solution to this problem relies on the presence of an orbital moment associated to the chirality, as recently pointed out in another context in [8]. This allows the qubit to be measured as if it had a spin, with the important difference, however, that a magnetic field cannot induce a rotation of the qubit. Note that a chiral doublet ground state has been recently reported in a dysprosium based molecular magnet [9]. For reasons explained below, we will discuss in detail an alternative implementation with Cu atoms on a surface.

We note that in the quantum information community, the coding of qubits in specific degrees of freedom has been investigated in the context of noiseless subsystems [10]. In particular, it has been shown that one can use certain degrees of freedom of three spins as a qubit, which will be isolated from certain types of noise [11]. In this regard, our proposal can be seen as a particularly simple realization of a noiseless subsystem, where the protected degree of freedom is identified with the physical chirality and the logical qubit is directly manipulated and measured without any encoding procedure.

The usual starting point to discuss triangular antiferromagnets is the Heisenberg model

$$H_{\mathrm{Heis}} = J_1 \vec{S}_1 \cdot \vec{S}_2 + J_2 \vec{S}_2 \cdot \vec{S}_3 + J_3 \vec{S}_3 \cdot \vec{S}_1 - g\mu_B \vec{H} \cdot \vec{S}, \quad (1)$$

where $\vec{S}$ is the total spin. When $J_1 = J_2 = J_3 = J$, the ground state is fourfold degenerate, and a convenient basis is provided by the simultaneous eigenstates of the scalar chirality $\vec{S}_1 \cdot (\vec{S}_2 \times \vec{S}_3)$ and of the projection S^α of the total spin in an arbitrary direction α:

$$|R, \sigma\rangle = (|-\sigma\sigma\sigma\rangle + \omega|\sigma - \sigma\sigma\rangle + \omega^2|\sigma\sigma - \sigma\rangle)/\sqrt{3}$$

$$|L, \sigma\rangle = (|-\sigma\sigma\sigma\rangle + \omega^2|\sigma - \sigma\sigma\rangle + \omega|\sigma\sigma - \sigma\rangle)/\sqrt{3},$$

where $\omega = \exp(2i\pi/3)$. $\sigma = \pm 1/2$ refers to S^α while L and R stand for left and right and refer to the chirality. To get a twofold qubit, we apply a positive magnetic field H in the direction α, which leaves us with a twofold degenerate ground state $\{|R, +1/2\rangle, |L, +1/2\rangle\}$ separated from the other states by an energy $g\mu_B H$. The two ground states

are only distinguished by the chirality and thus constitute a nonspin qubit (see Fig. 1 for a pictorial description of these states in the case where the magnetic field is in the x direction). This is different from the situation where non SU(2) invariant terms in the Hamiltonian such as Dzyaloshinskii-Moriya interactions lift the degeneracy, in which case the two ground states differ by the chirality and by the spin [7].

Let us first concentrate on the measurement problem. Since we have already used the magnetic field to create the qubit, it might seem hopeless to rely on the magnetic field to measure it. This is not the case, however. Indeed, let us turn to a more microscopic description of the system in terms of a three-site Hubbard model:

$$H_{\mathrm{Hub}} = -t\sum_{i=1}^{3}\sum_{\sigma}(c_{i\sigma}^{\dagger}c_{i+1\sigma}) + \mathrm{H.c.} + U\sum_{i+1}^{3} n_{i\uparrow}n_{i\downarrow}, \quad (2)$$

with implicit periodic boundary conditions. With three electrons, a canonical transformation maps this Hamiltonian onto the Heisenberg model with coupling $J = 4t^2/U$ to second order in t/U. However, as recently emphasized in [8], charge fluctuations are not completely suppressed, and an orbital current proportional to $\vec{S}_1 \cdot (\vec{S}_2 \times \vec{S}_3)$ can be obtained by inverting the canonical transformation. These orbital currents create an orbital moment perpendicular to the plane of the triangle, which can couple to an external magnetic field. Let us look at the magnitude of this orbital moment. In [8], this was calculated in perturbation theory and it was found that it is of order t^3/U^2, hence very small. However, having in mind atomic Cu, where the outer electron resides in the extended $4s$ orbital, we have calculated the orbital moment for an arbitrary value of the ratio t/U. The orbital moment is related to the magnetic field by $\mu_{\mathrm{orb}} = -\partial E/\partial B|_{B=0}$. The magnetic field is taken perpendicular to the plane of the triangle and is treated with the Peierls substitution as a phase: $t = |t|e^{i\varphi}$. The phase is such that its circulation is equal to the flux of the field, which leads to $\varphi = (\pi/2\sqrt{3})Ba^2/\phi_0$, where a is the intersite distance and $\phi_0 = hc/e$ is the flux quantum. Using all symmetries, the ground state energy can be obtained as

$$E = \rho\cos\left[\frac{1}{3}\arccos(A(B + C\sin 3\varphi)) + \frac{2\pi}{3}\right] + \frac{2U}{3}, \quad (3)$$

with $\rho = \sqrt{4U^2/9 + 12t^2}$, $A = -9/\rho(U^2 + 27t^2)$, $B = 2U^3/27$, and $C = 6\sqrt{3}t^3$. This leads to the following asymptotic expressions for the derivative:

$$-\frac{\partial E}{\partial \varphi} = \begin{cases} \sqrt{3}t(1 - U^2/6t^2), & U/t \ll 1, \\ 18\sqrt{3}t^3/U^2, & U/t \gg 1. \end{cases} \quad (4)$$

The full dependence as a function of U/t is depicted in Fig. 2. The moment is a relatively slowly decreasing function of U/t: at $U/t = 5$, it is still equal to about half its value at $U = 0$. To get an estimate of the actual magnitude of the moment, we use Harrisson's empirical dependence

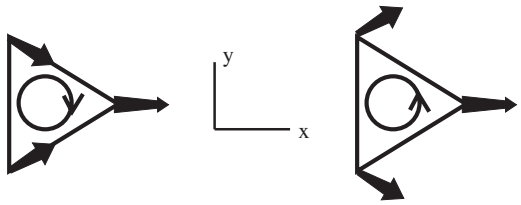

FIG. 1. Schematic view of the two chiral states. The two chiralities are illustrated by their classical analogs, namely, spin configurations forming umbrellalike structures with a net moment along the x axis, and with opposite chiralities. They are associated with currents circulating in opposite directions.

of the hopping integral on the intersite distance [12] relevant for the $4s$ orbitals of Cu: $t(\mathrm{eV}) = 10.67/a(\text{Å})^2$. This leads to an orbital moment $\mu_{\mathrm{orb}} = 0.65\mu_B$ at $U = 0$, a surprisingly large value in view of the results of Ref. [8]. For realistic values of U/t, the orbital moment is thus expected to be a sizable fraction of a Bohr magneton (see Fig. 2). So one can use a field parallel to the plane of the triangle to create the twofold degenerate ground state, and measure the chirality by inducing a component of the field perpendicular to the cluster to lift the degeneracy.

This effect will presumably be hard to detect in currently available Cu_3 clusters for two reasons: first of all, the orbital moment depends on the magnitude of the intersite hopping, which will be much smaller for $3d$ electrons than for $4s$ electrons, as assumed in the above estimate. Besides, if the qubit is of mixed character due to spin-orbit coupling, an external magnetic field will couple to both the orbital moment and the total spin, and the spin moment will dominate the splitting.

We thus turn to a description of an alternative implementation of such a qubit (see Fig. 3 for a representation of a chain of such qubits). The basic idea is to work with atomic Cu deposited on a surface rather than with molecules with Cu^{2+} ions. The main advantages are (i) the absence of spin-orbit coupling since the electron that carries the spin is in a $4s$ orbital, and (ii) the extension of the $4s$ orbitals, which leads to a larger ratio t/U. The imposi-

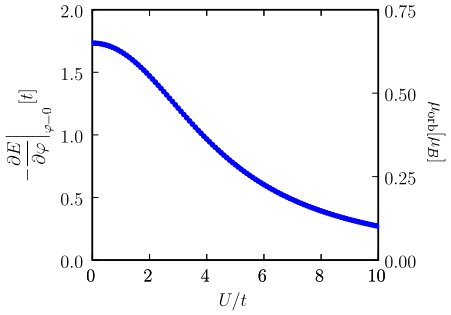

FIG. 2 (color online). Orbital moment associated to the chirality as a function of the ratio U/t. Right scale: in units of Bohr magneton, assuming the empirical dependence $t(\mathrm{eV}) = 10.67/a(\text{Å})^2$ for Cu $4s$ orbitals, where a is the intersite distance. Left scale: Opposite of the derivative of the energy with the phase calculated at $\varphi = 0$ in units of t.

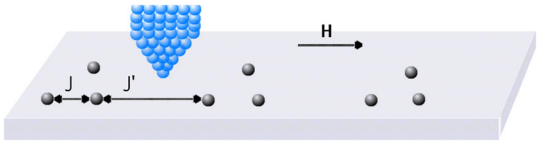

FIG. 3 (color online). An example of chirality-based qubit chain: triangular clusters of Cu atoms deposited on a surface. J is the interaction strength between spins inside one triangle. Its spatial modulation induced by approaching an STM tip of a triangle controls the one-qubit gates (see text). J' is the interaction strength between closest spins of two neighboring triangles induced by approaching an STM tip. It controls the two-qubit gates (see text). H is the in-plane magnetic field.

tion of a magnetic field perpendicular to the plane of the triangles will align the qubit towards a state whose orbital moment is polarized in the direction of the field. This allows one to prepare the qubit in a given chirality. Positioning of atoms in well-defined arrays with precision below the Angström level has been routinely realized using STM tips to move and place atoms (see, e.g., [13] for a review). Equilateral triangular arrays are especially easy to build taking advantage of the crystalline structure of the atoms of the substrate.

To use this system as a qubit, one should be able to manipulate this degree of freedom. To create one-qubit gates and modify the state of one qubit, one can modify the couplings in a controlled way by use of STM tips (in a way which has some similarity with the ones proposed in [7,14]). The control can be achieved by calibrating the system, performing repeated preliminary modifications of the couplings, and measuring the result. If the couplings are transformed from $J_1 = J_2 = J_3 = J$ to $J_1 = J + \delta J_1$, $J_2 = J + \delta J_2$, $J_3 = J + \delta J_3$, the states $|L\rangle$ and $|R\rangle$ are not any more eigenstates of (1). The Hamiltonian (1) in the basis $(|L\rangle, |R\rangle$ reads

$$H = \begin{pmatrix} -D - 3J/4 & V_1 + iV_2 \\ V_1 - iV_2 & -D - 3J/4 \end{pmatrix}, \qquad (5)$$

where the real parameters D, V_1, and V_2 are defined by $D = -(\delta J_1 + \delta J_2 + \delta J_3)/4$ and $V_1 + iV_2 = (\omega \delta J_1 + \delta J_2 + \omega^2 \delta J_3)/2$. The evolution operator through the perturbation is therefore $U = \exp -i(DI + V_1\tau^x - V_2\tau^y)t = \exp(-iDt)\exp -i(V_1\tau^x - V_2\tau^y)t$, where I is the identity operator and τ^x and τ^y are Pauli matrices associated with the chirality. By an appropriate choice of the three parameters δJ_1, δJ_2, δJ_3 and of the time of evolution t, it is therefore possible to engineer any rotation of the qubit of axis x or y. It is known that any one-qubit gate can be written [1] as $\exp(i\alpha)R_y(\beta)R_x(\gamma)R_y(\delta)$, where $R_y(\theta)$ [respectively $R_x(\theta)$] is the rotation of angle θ around the y axis (respectively x axis), and α, β, γ, δ are real parameters. Thus one can generate any one-qubit gate by a sequence of well chosen modifications of the couplings inside the triangle.

To perform a two-qubit gate, one should couple two neighboring triangles. If the triangles are sufficiently far apart, the permanent coupling between the spins of different triangles can be made small enough to be negligible. However, if they are not too far apart, a STM tip approached between the closest spins of two neighboring triangles can mediate an interaction between two spins. There is obviously an optimal distance between the triangles to respect in the best possible way these two requirements. In the presence of the STM tip, the new interaction J' between the closest spins of each triangle creates an interaction between the two triangles i and j which, in the basis of the qubits, takes the form

$$H' = (J'/9)\vec{S}_i \cdot \vec{S}_j(1 + 2\tau_i^x + 2\sqrt{3}\tau_i^y)(1 - 4\tau_j^x), \qquad (6)$$

where $\vec{S}_i$ and $\vec{S}_j$ are the total spins of the triangles [15]. Since the spins are polarized by the magnetic field parallel to the surface, we can consider $\vec{S}_i \cdot \vec{S}_j = 1/4$ to be constant. Since an arbitrary one-qubit gate can be made on each triangle, this can be transformed by rotations of the qubits to $(J'/36)(1 - 4\tau_i^z)(1 - 4\tau_j^z)$. The evolution operator through this interaction is up to one-qubit gates $\exp(-i(4J'/9)\tau_i^z\tau_j^z t)$; if applied for a time $t = 9\pi/(16J')$ it generates the two-qubit gate $\exp(-i\pi/4)\mathrm{diag}(1, i, i, 1)$ in the basis $(|00\rangle, |01\rangle, |10\rangle, |11\rangle)$, equivalent up to one-qubit operations to a CNOT gate [16]. Since the CNOT gate plus one-qubit gates form a universal set [17], the model proposed can generate any arbitrary sequence of gates.

The interaction time necessary to implement the one- and two-qubit gates is $t_g \propto 1/\delta J$, where δJ is the typical magnitude of the process $[|\omega\delta J_1 + \delta J_2 + \omega^2\delta J_3|$ in Eq. (5), $\delta J'$ in Eq. (6)]. If the tip is mechanically moved, time scales reached in experiments of the early 1990s were of the order of 10 ns [18]. This time can be significantly reduced, down to the picosecond range, by using a nonmoving photosensible tip optically addressed [19]. The range 10 ps $< t_g <$ 10 ns corresponds to 0.004 K $< \delta J <$ 4 K. With a coupling J of the order of 100 K, typical for kinetic exchange, this corresponds to small, hence reasonable, modifications of the exchange integrals that can be induced by the influence of the tip. Note that the three important steps in the manipulation of a qubit can be optimized independently since they depend on different parameters: the orbital moment used to initialize and measure the qubit depends on t and U in (2) and is thus related to J in (1), one-qubit gates depend on the modulations δJ_i, and two-qubit gates are controlled by the coupling J' induced between the qubits. This should help to tune the system to the optimal working point. We also note that the system operated this way should be scalable when increasing the number of qubits: indeed, the qubits are individually addressed and well separated spatially.

Finally, let us discuss the problem of decoherence and why we think such a qubit is well protected. There are *a priori* two sources of decoherence: local vibrations, and magnetic noise. Local vibrations can be a source of decoherence because they change the bond lengths inside the Cu_3 cluster, hence the exchange integrals. As usual, zero-

point vibrations are not a source of decoherence. In the present case, this is easy to see since, as long as the cluster has a C_3 axis, the ground state for a given total spin is twofold degenerate, even if spin-phonon coupling is included. Indeed the degeneracy of the ground state on which the qubit is based comes ultimately from the fact that the C_3 group has a two-dimensional irreducible representation to which the ground state belongs, and this will remain true for the system including phonons.

Regarding thermal fluctuations, the main mechanism of decoherence comes from the vibrations of individual Cu atoms in the potential well in which they are located. The depth of the well varies from one system to the other, but typical values of the energy barriers for diffusion are a fraction of an eV [20]. The corresponding frequencies of harmonic vibrations are in the terahertz range, i.e., 50 K. So, by working at low enough temperature, these thermal vibrations can be exponentially suppressed. Since subkelvin temperatures are now accessible to STM experiments, it should be possible to reduce this source of decoherence very efficiently. The other mechanism is related to vibrations of the substrate, or phonons. We believe that our qubit is naturally protected against these vibrations. Indeed, at low temperature, only long-wave length acoustic modes can be excited, and they are essentially decoupled from chirality since it is only sensitive to local differences in bond lengths inside the Cu_3 cluster.

The other source of decoherence is magnetic noise. The specificity of the qubit proposed in the present Letter is that the chirality is not coupled to a uniform field, as stated before. So decoherence can only be induced by fluctuating fields that are inhomogeneous on the scale of the Cu_3 cluster. A simple calculation similar to the derivation of Eq. (6) shows that the transition amplitude between the two chirality states is equal to $-(1/3)(h_1^\alpha + \omega^2 h_2^\alpha + \omega h_3^\alpha)$, where α is the direction of the field used to fix the orientation of the total spin parallel to the plane, and $\vec{h}_i$ is the local field at site i of the cluster. Now, local spins such as nuclear spins in the neighborhood of the cluster will produce inhomogeneous fields. The typical decoherence time can be expected to be similar to that observed in nanomagnets, where such a mechanism dominates, hence to be of the order of the microsecond [21]. However, by choosing a substrate with little or no nuclear spins, one could get rid of this effect altogether. Interesting possibilities are silicon based substrates such as silicon itself [22] or SiO_2, where the only nuclear spins are carried by ^{29}Si, of natural abundance 4.7%, or carbon based substrates such as graphene, with nuclear spins on ^{13}C only, of natural abundance 1%. A few percent of nuclear spins is probably acceptable since spins far from the cluster will produce an almost homogeneous field, but in any case isotopic purification could be used to reduce further this channel of decoherence.

So, working at low enough temperature and with the appropriate substrate should allow one to reach decoherence times of several microseconds, much longer than the operation time, which is in the nanosecond-picosecond range.

In conclusion, we have shown how to build a nonspin qubit out of a magnetic cluster. The two states of a qubit are the lowest energy states of opposite chirality of an equilateral triangular cluster of atoms with an in-plane magnetic field. We have shown that electric fields generated by STM tips are sufficient to create a universal set of quantum gates. The presence of an orbital magnetic moment perpendicular to the plane of the triangle enables one to prepare the system and then to measure it at the end. The structure of the chiral states makes them robust against noise, and decoherence times should be long enough to perform many gates using current technology. We thus think our proposal has several attractive features for qubit implementation.

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