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Rabi oscillations of pinned solitons in spin chains: A route to quantum computation and communication.

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We provide the first evidence for coherence and Rabi oscillations of spin-solitons pinned by the local breaking of translational symmetry in isotropic Heisenberg chains (simple antiferromagnetic-Néel or spin-Peierls). We show that these correlated spin systems made of hundreds of coupled spin bear an overall spin $S=1/2$ and can be manipulated as a single spin. This is clearly contrary to all known spin-qubits which are paramagnetic centres, highly diluted to prevent decoherence. These results offer an alternative approach for spin-qubits paving the way for the implementation of a new type of quantum computer.

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Most physical, chemical or biological systems showing quantum oscillations are of relatively small size: isolated NV-centers in diamond¹, $4f$ or $3d$ transition-metal ions (single-spins, 0.1 nm)^{2,3}, single molecule magnets⁴ (15 spins, 1 nm), or marine algae (5 nm wide proteins)⁵. Their environmental couplings are necessarily weak in order to reduce damping⁶. In magnetic systems, decoherence is usually dominated by spin-bath dipole-dipole interactions⁷ and observations of quantum oscillations require *qubits dilution*. Here, we report the first experimental realization of what we call soliton qubits. Contrary to all existing qubits, each qubit is made of hundreds of strongly exchange-coupled spins sitting at defects of strongly correlated spin-chains. This takes advantage of one of the most remarkable properties of one-dimensional spin systems: their quite unconventional response to translational symmetry breakings which often consists in the formation of magnetic defects described as spin solitons (kinks or domain walls). In as grown single-crystals, these defects are generally associated with local inhomogeneities such as crystallographic defects/disorder, bond alternations, chain ends etc. Theoretical prediction for Heisenberg spin 1/2 quantum spin chains show that each of these magnetic defects carry an overall spin $S=1/2$ which can serve as a qubit. The idea of pinned- soliton was successfully applied to explain the magnetic susceptibility⁸, NMR⁹ and some EPR experiments¹⁰ in spin chains, however, to date almost *nothing is known either experimentally or theoretically about the coherent dynamics of solitons*.

In this communication, we report for the first time on Rabi oscillations of pinned spin-solitons in isotropic Heisenberg chains (simple antiferromagnetic-Néel or spin-Peierls). We demonstrate that these collective extended defects bear an overall spin $S=1/2$ and can be manipulated as single quantum spins. This clearly presents an alternative to all known spin-qubits which are paramagnetic centres in highly diluted samples. These results offer a new approach for spin-qubits paving the way for

the implementation of a new type of quantum computer.

The first evidence of Rabi oscillations in sol-qubits is obtained on single-crystals of the so-called antiferromagnetic quantum spin chains $(\text{TMTTF})_2\text{X}$, with $\text{X} = \text{AsF}_6$, PF_6 , SbF_6 (Fig. 1). This family of organic magnets, also called Fabre salts¹¹, was extensively studied during the last decades and shows an extremely rich phase diagram¹². The systems with $\text{X} = \text{AsF}_6$ and PF_6 show a gapped dimerized spin-pair singlet ground-state below their spin-Peierls transitions at $T_{SP} = 13$ K and 19 K respectively, whereas the system with $\text{X} = \text{SbF}_6$ exhibits a Néel antiferromagnetic phase below $T_N = 7$ K.

The Hamiltonian of a $S=1/2$ Heisenberg chain can be written:

$$H = J \sum_i [(1 - \delta)S_{2i-1}S_{2i} + (1 + \delta)S_{2i}S_{2i+1}] \quad (1)$$

where S_i are the $S=1/2$ spin operators, J the exchange coupling and δ the dimerization parameter. If $\delta = 0$, (1) describes the uniform Heisenberg AF chain the ground state is a gapless $S=1/2$ doublet. This is the case of the spin-chain system $(\text{TMTTF})_2\text{SbF}_6$ at $T > T_N$. If $\delta > 0$, (1) describes a Spin-Peierls chain: the ground state is a gapped dimerized spin-pair singlet ($S=0$) at temperatures below T_{SP} . This is the case of the systems $(\text{TMTTF})_2\text{PF}_6$ and $(\text{TMTTF})_2\text{AsF}_6$ (if $T > T_{SP}$, $\delta = 0$ in these systems too). The isotropic part of the exchange interaction of these three systems, $J \sim 400$ K, is relatively large whereas their intra- and inter-dimer contributions of $(\text{TMTTF})_2\text{PF}_6$ and $(\text{TMTTF})_2\text{AsF}_6$, give a bond alternation (dimerization) parameter $\delta \sim 0.03$ leading to the singlet-triplet gap $\Delta = 35$ K¹⁴. Such a value is more than enough to provide an excellent separation of the ground state at the Kelvin scale of temperatures, i.e. an extended collective singlet ground-state in which two trapped soliton qubits can be strongly entangled.

The single crystals of $(\text{TMTTF})_2\text{X}$ were grown by an electrochemical technique¹⁵. The crystals are needle-shaped with typical dimensions: $3 \times 0.5 \times 0.1$ mm³. They

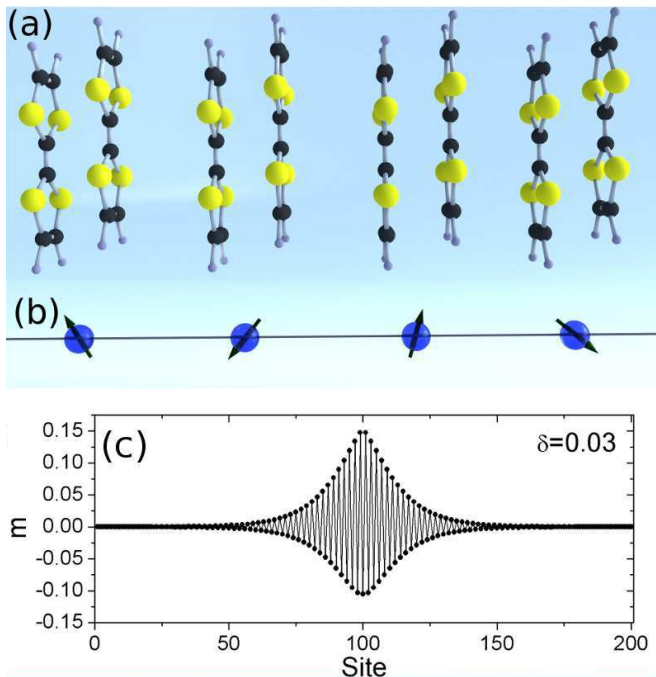


FIG. 1. Colour online. (a) Schematic representation of $(\text{TMTTF})_2\text{X}$ chain. The TMTTF molecules are stacked in the a axis direction forming the chain. Each double molecules of TMTTF carries a spin $S = 1/2$. The counter anion X is not represented here. (b), magnetic representation of the chain. The blue spheres are the double molecules of TMTTF carrying a spin (black arrow) and coupled along the a axis by the exchange $J(1 \pm \delta)$. (c), site magnetization profile induced by one bond defect placed in the middle of dimerized chain in the $M_z = 1/2$ space computed by DMRG (see method¹³).

crystallize in the triclinic $\text{P}\bar{1}$ space group. The magnetic principal axes (b' and c^*) are different from the crystallographic axes and correspond to the extrema of g -factor in the plan perpendicular to a axis. The static magnetic field can be applied in any direction in the $b'-c^*$ plane. For each set of measurements a fresh sample was used.

Continuous Wave (CW) and Pulsed Electron Paramagnetic Resonance experiments were performed with the three systems using a conventional X-band Bruker spectrometer operating at about 9.6 GHz between 3 K and 300 K and enabling sample rotations. The crystals were glued on the sample holder with their a -axis oriented along the microwave field (h_{mw}) direction which is the same as the sample-rotation direction (the static H being applied in the basal $b'-c^*$ plane with θ the angle between H and c^*).

Above 30 K a single Lorentzian-shaped EPR line (main line) is observed, displaying an anisotropy of g factor, associated with different orientations of H . This anisotropy and the temperature dependence are typical of uniform quantum Heisenberg spin chains and was intensively studied in the past¹⁶ Below about 30 K a second EPR line, a very sharp one, appears in the three systems at the same magnetic field as the main line. The integrated

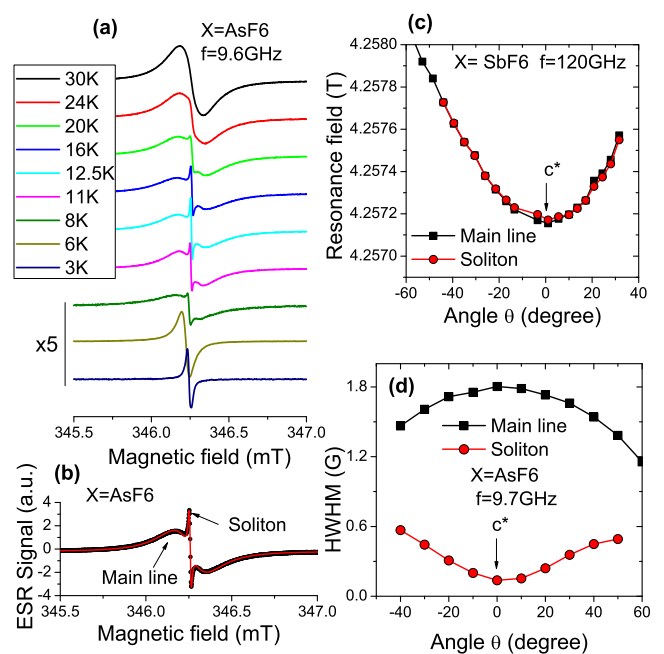


FIG. 2. color online. CW-EPR, evidence of the soliton signal. (a) Set of CW-EPR spectra of $(\text{TMTTF})_2\text{AsF}_6$ when the temperature decreases for $H \parallel c^*$. (b) EPR spectrum of $(\text{TMTTF})_2\text{AsF}_6$ at $T = 12.5$ K. The dots are the experiments and the red line is a fit of 2 derivative of Lorentzian. (c) Angular dependence of the resonance fields of $(\text{TMTTF})_2\text{SbF}_6$ at 120 GHz. The black squares are the main lines, the red circles are the soliton lines. (d) Angular dependence of the linewidth of $(\text{TMTTF})_2\text{AsF}_6$ at 9.5 GHz and $T = 12.5$ K.

intensity of this sharp signal (SS) is much smaller (by a factor of $10^2 - 10^3$) than the one of the main line, indicating its defect origin.

As an example, figure 2(a) gives a set of spectra obtained with $\text{X} = \text{AsF}_6$ between 30 K and 3 K with $H \parallel c^*$ showing how the very sharp signal progressively appears and becomes dominant when the temperature decreases. Below $T_{SP} = 13$ K the system starts to dimerise and enters in the spin-Peierls phase. The intensity of the main peak drops but that of the SS remains almost unchanged. The same behaviour has been observed in PF_6 compound. In the SbF_6 system, the linewidth of the main line diverges when T decreases down to $T_N = 7$ K whereas the SS remains almost unchanged, except below T_N where it disappears¹⁷. This behaviour is contrary to that of an isolated paramagnetic impurity, the intensity of which is strongly temperature-dependent (Curie law), and is rather characteristic of an ensemble of correlated spins.

In the limit of the resolution of the X-band spectrometer, the measured resonance field and therefore the g factor were identical for both the broad and sharp peaks and did not change significantly for different molecules. A two-lorentzian fit shows that their linewidths differ by a factor of ten (~ 1 G for the former and ~ 0.1 G for

the latter, Fig. 2(b)). In order to improve the resolution we measured the CW-EPR in these systems at 120 GHz in a home-made quasi-optical CW-EPR spectrometer¹⁸ (the angular dependence of the magnetic field resonance of the SbF_6 system is given in figure 2(c)). The resonance fields of both lines remain identical for all the applied-field orientations, to an accuracy better than 10^{-5} .

Figure 2(d) gives the linewidth angular dependence of the two peaks observed. The broad/sharp linewidth ratio reaches its maximum of ~ 10 when the static field is applied along the c^* axis. Surprisingly, whereas the resonant field of the two peaks is observed to be the same whatever the angle θ , the angular dependences of their linewidths are opposite. The width of the broad line follows the well-known behavior of a uniform $S = 1/2$ chain, proportional to $1 + \cos^2 \theta$, with a maximum for $\theta = 0^\circ$, whereas the one of the sharp line is minimum for $\theta = 0^\circ$ as expected for a line issued from defect-induced correlated spins.

To sum up, while the SS has quite unusual, compared to a isolated impurity signal, the temperature and the linewidth angular dependencies, one of the most important features which supports its spin soliton origin is the finding that SS g-factor is *precisely the same as the one of the main EPR line*. This result is perfectly consistent with a physical picture which one can suggest to describe the defect formation in TMTTF charge transfer salts. As we have mentioned above any translation symmetry breaking of molecular structure in the direction of the chain, crystallographic or electron charge inhomogeneities, will lead locally to intra-chain exchange modification which in turn will cause a spin soliton formation close to this defect. The observed EPR signal in this case will come not from an impurity centre itself (a localized electron somewhere in TMTTF molecule) but from a collective precession of hundreds of non perturbed spins of the chain which form a soliton. Quite naturally this precession is characterized by the same g-factor as the main EPR signal.

Before going further in our experimental investigation, let us explain in more details the condition of appearance and stability of solitons near bond defects in spin chains. For that we have performed numerical DMRG studies of alternating spin chains in which the successive bonds are $J_1 = J(1+\delta)$, $J_2 = J(1-\delta)$ with $J_1 > J_2$. In fig. 1c a typical distribution of the soliton magnetic moment is shown for a defect characterized by the succession of two strong bonds placed in the middle of the chain. The result, shown in Fig 1, fully corresponds to a spin-soliton, and after integration we find that it carries a $S = 1/2$. These results are in full agreement with similar studies previously published¹⁹ and²⁰ for comprehensive theoretical studies.

The pulsed EPR experiments were performed on $(\text{TMTTF})_2\text{AsF}_6$ and $(\text{TMTTF})_2\text{PF}_6$ single crystals with a microwave field h_{mw} varying between 0.1 and 1.5 mT. The coherent signal, resulting from the SS observed in CW experiments, was recorded by the Free Induc-

tion Decay method. The spin-echo detection cannot be used here, in contrast with most other known systems, because of the absence of sizable inhomogeneous line-broadening. In fact the line-broadening of ~ 0.1 G observed is essentially homogeneous (see discussion below). Examples of Rabi oscillations obtained at 3 K for $X = \text{PF}_6$ are given in figure 3. The oscillations are very well-fitted by the exponentially damped sinusoidal function $\langle S_x(t) \rangle \propto \sin(\Omega_R t) \exp(-t/\tau_R)$ with Ω_R the Rabi pulsation and τ_R the Rabi damping characteristic time. The Rabi frequency increases linearly with h_{mw} with a slope $d(\Omega_R/2\pi)/dh_{mw} \sim 28$ MHz/mT close to the expected value for spins $S = 1/2$ (Fig. 3(b)), thus providing an additional evidence in favour of our model of a trapped soliton with $S = 1/2$. This figure also shows the microwave-field dependence of the Rabi damping $1/\tau_R$ allowing one to evaluate the damping by the microwaves^{2,4,21}. This “over-damping”, associated with inhomogeneous line-broadening due to the distributions of the transverse g factor (resulting itself from weak ligand-field distributions) or to the microwave-field amplitude (non-homogeneous cavities) is generally unavoidable²². In the present case, and this is rather exceptional, the “over-damping” is particularly small due to the homogeneous character of the EPR line. The measured value of $d(1/\tau_R)/dh_{mw} = 0.4$ MHz/mT (Fig. 3(b)), is 10 to 50 times smaller than in ion-diluted systems^{21,22}. In addition, contrary to most other systems, the figure of merit $Q_M = \Omega_R \tau_R / 2\pi$ of sol-qubits in $(\text{TMTTF})_2\text{PF}_6$ does not saturate when the Rabi frequency increases (Fig. 3(c)) and follows the expression $Q_M = \Omega_R / (8.5 + 0.015\Omega_R)$. Our largest microwave field $h_{mw} = 1.5$ mT gives at 3 K $Q_M \sim 23$ while a value of the order of 70 is expected for a larger field. Whereas Q_M of $\text{CaWO}_4:\text{Er}^{3+}$ and $\text{MgO}:\text{Mn}^{2+}$ which saturate at about 3, Q_M of sol-qubits is larger by an order of magnitude.

The fact that sol-qubits are almost not sensitive to system parameters (g factors, inhomogeneous magnetic and microwave fields, etc.) is worthy of special consideration. Indeed, with all the other types of qubits these distributions lead to inhomogeneous distributions of Rabi frequencies (the qubits are not identical) giving destructive interferences and decoherence. This is a major roadblock for the implementation of a spin-based quantum computer. Magnetic dipole-dipole interactions are also an inevitable and constitute a source of decoherence unless the qubits are very far from each others preventing any kind of manipulation. With sol-qubits, the situation is just opposite: the strong spin exchange interactions ($J=400$ K) eliminates decoherence through the well known exchange narrowing mechanism, as this is shown in the $S=1$ Haldane spin chains²³ picture developed for interacting $S=1/2$ degrees of freedom. This explains why the coherence of sol-qubits is robust against microwaves even at high power. Finally these long-living sol-qubits, even if they are distant, are easily coupled to each others and controlled through effective isotropic exchange interaction along the spin-chain.

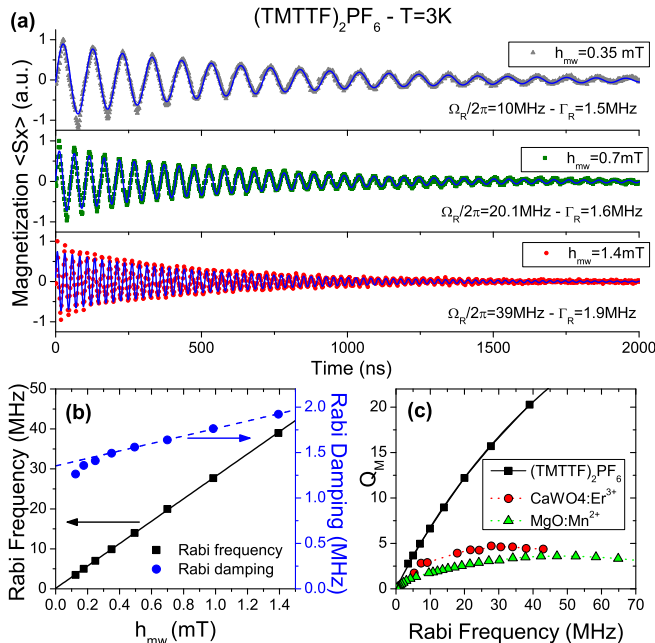


FIG. 3. colour online. Rabi oscillations and coherence properties of soliton-qubit. (a) Series of Rabi oscillations of $(\text{TMTTF})_2\text{PF}_6$ measured at $T = 3$ K and $f = 9.7$ GHz using the FID method while increasing the microwave magnetic field h_{mw} . Each point is an average of 1000 FID measurements. The blue lines are fits using $\sin(\Omega_R) \cdot \exp(t/\tau_R)$. (b) Rabi frequencies ($\Omega_R/2\pi$) and damping ($1/\tau_R$) as function of h_{mw} . The black line represents the Rabi frequency of spin $S=1/2$. The blue line is a fit using $\Gamma_0 + \gamma \cdot h_{mw}$, $\Gamma_0 = 1.35$ MHz (the zeros microwave field coherence) and $\gamma = 0.41$ MHz/mT the effect of microwave field on the Rabi damping. (c) Merit factor Q_M of $(\text{TMTTF})_2\text{PF}_6$ compared to diluted ion systems²². The black line is a simulation using the Γ_0 and γ from (b).

In conclusion, by observing long-living Rabi oscillations of sol-qubits in Heisenberg gapped spin-Peierls systems, we provide first evidence for coherence in spin chains and more particularly in solitons trapped at exchange defects in spin-chains. Due to isotropic inter-qubit exchange interaction, the EPR lines observed are homogeneous and narrowed, eliminating all the usual decoherence mechanisms such as the one associated with non-perfectly identical qubits and dipolar interactions.

Following the idea of spin-qubit quantum computer²⁴, an increasing number of proposals were made during the last decades showing theoretically how spin chains may enable the implementation of a quantum computer by using them as quantum wires to connect distant qubit registers without resorting to optics^{25–30}. We speculate that our sol-qubits might be ideal candidates for the realization of such a computer since they represent intrinsic spin registers which do not require the addition of any supplementary spin to the system and since they perfectly match the communication channel - spin chain.

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