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Quantum coherence of strongly correlated defects in spin chains.

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Abstract

Most of qubit systems known to date are isolated paramagnetic centres in magnetically diluted samples since their dilution allows to considerably weaken the dipole-dipole inter-qubit interaction and thus to prevent the decoherence. Here we suggest an alternative approach for spin qubits which are built on spin $S = 1/2$ defects in magnetically concentrated strongly correlated systems - spin chains. The corresponding qubits are made of spin solitons resulting from local breaking of transitional symmetry associated with point-defects. We provide the first evidence for coherence and Rabi oscillations of spin solitons in isotropic Heisenberg chains, simple antiferromagnetic-Néel or spin-Peierls, proving that they can be manipulated as single spin $S = 1/2$. The entanglement of these many-body soliton states over macroscopic distances along chains gives rise to networks of coupled qubits which could easily be decoupled at will in extensions of this work.

Keywords: Quantum coherence, EPR, strongly correlated magnet

1 Introduction

Most physical, chemical or biological systems showing quantum oscillations are of relatively small size: isolated NV-centers in diamond [1], $4f$ or $3d$ transition-metal ions (single-spins, 0.1 nm) [2, 3], single molecule magnets [4] (15 spins, 1 nm), or marine algae (5 nm wide proteins) [5]. Their environmental couplings are necessarily weak in order to reduce damping [6]. In magnetic systems, decoherence is usually dominated by spin-bath dipole-dipole interactions [7] and observations of quantum oscillations require *qubits dilution*. Here, we demonstrate the feasibility of a new approach for magnetic qubits, called soliton-qubit or sol-qubit [8], which relies on spin $S = 1/2$ defects in *magnetically concentrated* strongly correlated systems

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- spin chains. An increasing number of proposals were made during the last decades showing theoretically how spin chains may enable the implementation of a quantum computer. Most of them discuss the possibility of using spin chains as quantum wires to connect distant qubits registers without resorting to optics [9, 10, 11, 12, 13, 14, 15]. It was concluded that, in spite of short-ranged spin correlations, most spin-chain models exhibit the long-distance entanglement [10, 12, 14], thus a spin chain can be used as a communication channel. However, according to this scheme, to realize a qubit network one needs first to create qubits and then to match them to the communication channels.

From experimental side, however, the situation is less clear. While there are some experimental indications that the macroscopic entanglement can be achieved in spin systems [15], almost nothing is known about quantum dynamics of sol-qubits. Moreover, the general experimental situation concerning the coherent dynamics of magnetic cluster, nanomagnets, rings etc rather indicates that the decoherence time in these objects is very short thus preventing their use as qubits.

This study of sol-qubits was performed on single-crystals of the so-called antiferromagnetic quantum spin chains $(TMTTF)_2X$, with X = AsF_6, PF_6, SbF_6 (Fig. 1). This family of organic magnets, also called Fabre salts [16], was extensively studied during the last decades and shows an extremely rich phase diagram [17]. The physical properties of these magnets depend on the counter anion X of the molecule and are found to be quite sensitive on the presence of paramagnetic defects (spin-Peierls and superconducting transition). The systems with X = 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 X = SbF_6 exhibits a Néel antiferromagnetic phase below $T_N = 7$ K.

2 Experimental details

The single crystals of $(TMTTF)_2X$ were grown by an electrochemical technique. The crystals are needle-shaped with typical dimensions: 3x0.5x0.1 mm^3. The samples crystallize in the triclinic $P\bar{1}$ space group. The magnetic principal axes (b' and c^*) are different from the crystallographic axes [18]. 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 9.7 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). The pulsed EPR experiments were performed on $(TMTTF)_2AsF_6$ and $(TMTTF)_2PF_6$ single crystals with a microwave field h_{mw} varying between 0.1 and 1.5 mT. The coherent signal, resulting from the sharp EPR line observed in CW experiments, was recorded by the Free Induction Decay method.

3 Results and discussion.

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 and a temperature dependence which are typical of uniform quantum Heisenberg spin chains [19]. Below about 30 K a second EPR line, a very sharp one, appears in the three systems at the same magnetic field

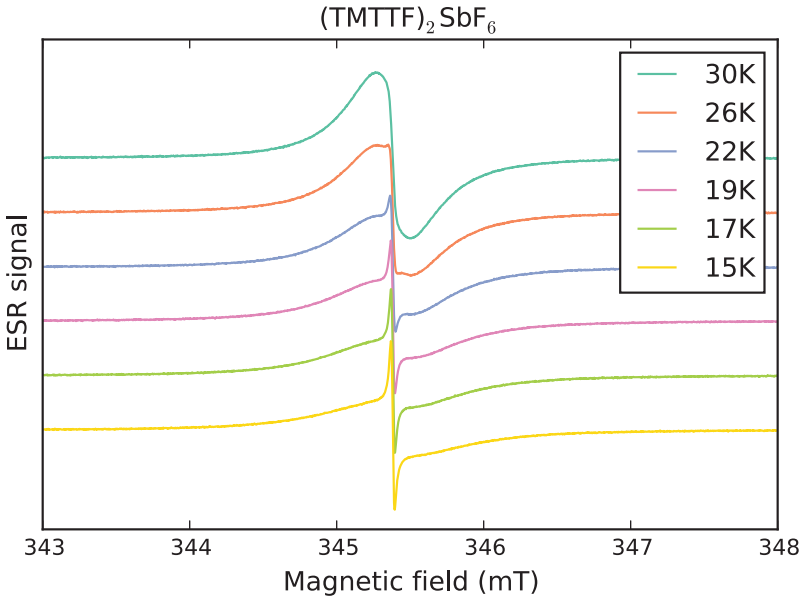

Figure 1: CW-EPR signal $(TMTTF)_2SbF_6$ recoder for H$\|$ c^* for temperature between T=30 K and 15 K. Below T=30 K a very sharp line is superimposed to the signal from the spin chain. The sharp signal comes from the strongly correlated defects.

as the one associated with the main line. The integrated intensity of this sharp signal is much smaller than the one of the main line, indicating its defect origin. The figure 1 shows an exemple of CW EPR signals recorded on $(TMTTF)_2SbF_6$ from 30 K down to 15 K. 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.

The temperature behaviour of EPR intensity of $(TMTTF)_2SbF_6$ and $(TMTTF)_2AsF_6$ is given in figure 2. χ_{ESR} of the main signal is quiet independent of the temperature for the three systems. For X=AsF_6 and PF_6 compounds, when $T < T_{sp}$ the intensity of the main line drops as expected in spin Peierls system. In The case of $(TMTTF)_2SbF_6$, the system enters in the Néel state at about 7 K and χ_{esr} of the main slightly decrease. We can conclude the main line is the signal from the Heisenberg spin chain. The signal from the sharp line is more intriguing. The intensity of the signal is mostly temperature independent. In paramagnetic impurities the intensity of the signal should follow a Curie law ($\chi \sim 1/T$), whereas here the intensity slightly decrease with temperature denoting a strong correlation to the spin chain. It has been shown [20] that quantitative measurement of correlated defects in quantum spin chains is not trivial. However we can estimate the concentration of defects in order of 10^{-2}-10^{-3}. Although the nature of the defects remain unclear, we think there are due to stacking faults during the crystallization process.

The Rabi oscillations measurements on the sharp line is given in figure 3. Each oscillation is a coherent inversion of the population of the correlated defects. On a contrary to diluted paramagnetic defects, the Rabi damping is almost independent of the microwave power [8]. In

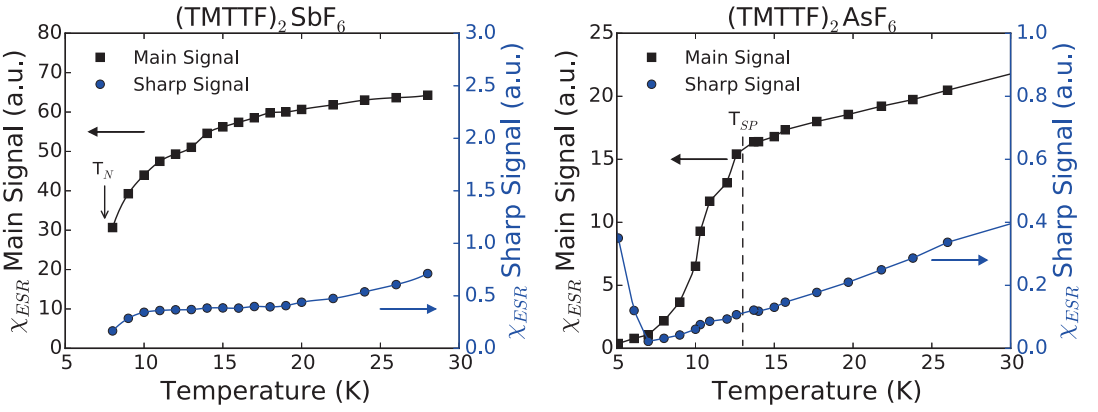

Figure 2: Intensity of CW EPR signals of $(TMTTF)_2SbF_6$ (left) and $(TMTTF)_2AsF_6$ (right). The Black squares and blue circles are intensities the main line and sharp line respectively.

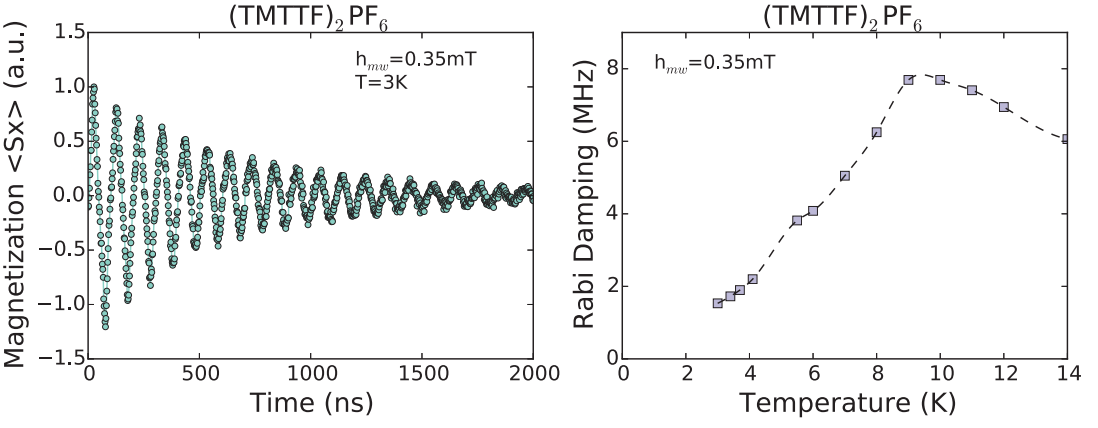

Figure 3: (left) Rabi oscillation of the strongly correlated defect observed in $(TMTTF)_2PF_6$ at T=3 K.(right) Rabi damping obtained from the fits of Rabi oscillations of the pinned solitons for temperature below 14 K. Dashed line is a guide for the eyes.

paramagnetic impurities, the Rabi damping can be reduces by decreasing the temperature but reach a threshold due to inhomogeneity of the microwave [21]. In the pinned soliton qubit, the Rabi damping keeps decreasing when temperature go down. This behaviour proves that the dynamic of such correlated defects is dramatically different to paramagnetic impurities and its strongly correlated nature protects it to the environmental coherence of dipolar and nuclear spin bath.

The fact that sol-qubits are almost not sensitive to system parameters is worthy of special consideration. Qubits are actually never identical and in most qubit systems they are coupled by the *magnetic dipole-dipole interaction* which is inevitably a major source of decoherence. In the present case the situation is just opposite: the interactions between qubits eliminate the

decoherence caused by inhomogeneous distributions of Rabi frequencies. The reason is that we deal here with an ensemble of $S = 1/2$ defects (sol-qubits) in a strongly correlated spin-chain system. In consequence, the dynamics of two sol-qubits even distant from each other is not independent but controlled by an effective *isotropic exchange* interaction. We anticipate that the physical picture developed for the interacting $S = 1/2$ degrees of freedom in Haldane spin chains with $S = 1$ can be applied here [22]. This leads to a kind of narrowing mechanism which homogenizes the parameter distributions explaining why the coherence of sol-qubits, rather robust against microwaves, is limited by $2T_2^*$.

4 Conclusion

In conclusion, we suggest that $S = 1/2$ defects in strongly correlated spin-chain systems can be used for quantum information processing. This is in striking contrast with known spin-qubits systems where qubits are uncoupled or weakly coupled by an anisotropic interaction leading to decoherence. By observing long-living Rabi oscillations of sol-qubits in Heisenberg gapped spin-Peierls systems, we provide first and clear evidence for the coherence of solitons trapped at defects in spin-chains. Due to an isotropic inter-qubit exchange interaction, the EPR lines observed are homogeneous and narrowed. This eliminates most of the decoherence mechanisms associated with disorder i.e. with non-perfectly identical qubits. Remarkably, the important effect of decoherence by the microwaves which tends to burst out with the number of qubits is shown to be here negligible.

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