Entangled spin states in self-assembled monolayer systems

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We discuss the opportunity for creation of an entangled spin state in a self-assembled monolayer molecular system. A group of spin radicals can be transferred to the entangled state using resonant electromagnetic pulses. The magnetic dipole–dipole interaction within a group is taken into account while the same interaction between the groups is neglected. A high external magnetic field at low temperature provides the identical entangled states for all spin groups in a sample. We discuss how this entangled state can be measured in experiments using the macroscopic magnetization.

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I. INTRODUCTION

Entangled states of weakly interacting particles and even of macroscopic objects have attracted much attention recently.[1] Besides exhibiting unusual physical properties, these states are expected to be useful in quantum communication and quantum information processing.[2]

In this paper we discuss how to create and detect entangled states in very promising novel materials—self-assembled monolayer (SAM) molecular systems. The advantages of applying thermodynamically driven or directed self-assembly to create highly organized surface molecular assemblies is becoming increasingly appreciated[3] and the formation of SAMs using molecules with complex structures, such as highly conjugated oligomeric molecules, at metal surfaces has been reliably demonstrated (see, for example, Ref. 4). Some of these SAMs allow voltage control of the conductivity states resulting in new classes of electronic devices including switches and memory cells (see, for example, Ref. 5).

Unlike the main focus of research in this area that concentrates on the electric properties of organized two-dimensional arrays of organic molecules, we will explore the magnetic properties of SAMs.

II. SYSTEM UNDER CONSIDERATION

We consider an ensemble of ''three-spin groups.'' As an example, each group consists of three radicals, and each radical has an electron spin, 1/2. Stable free radicals with spin 1/2 have been known for many years (see, for example, Ref. 6). We assume that the distance between the spin radicals in a group is large enough to neglect exchange interactions between spins but small enough to provide strong dipole–dipole interaction between them. For our purposes the shift of the electron spin resonance (ESR) frequency due to the dipole–dipole interaction inside the groups must be greater than the ESR linewidth. The distance between the spin groups must be large enough, so that the interaction between the groups can be ignored.

If all conditions mentioned above are satisfied, one has an ensemble of noninteracting three-spin groups. Assume that a sample of SAM containing three-spin groups is placed into a high external magnetic field, B_0. If the temperature is small enough ($k_BT \ll \mu_B B_0$) almost all spins will be in their ground states. Thus, one can manipulate an ensemble of spin groups which have identical quantum spin states. One possible geometry of the system which we are considering below is shown in Fig. 1. The external magnetic field in Fig. 1 is nonuniform in the *y*-direction but uniform in the *x*-direction, so that all spin groups experience the same conditions. We assume that the magnetic field difference between neighboring spin radicals allows one to address resonant electromagnetic pulses to a particular spin in the spin group. A large field gradient inside the group can be produced, for example, by micropattened wires shown in Fig. 1. As an example, 1 μm diameter wires with equal and parallel currents of 1 A can produce the magnetic field gradient of about 10^6 T/m, if the distance between wires is about 1 μm.[7]

III. ESTIMATES

Assume that the temperature is $T=1$ K, and the external magnetic field, is $B_0 \approx 10$ T. In this case, the radical spins are in their ground state. [The probability for an electron spin to change its direction is $\exp(-2\mu_B B_0/k_B T) \approx 1.4\times 10^{-6}$.] The ESR frequency, $f_e = \gamma_e B_0 \approx 280$ GHz (where γ_e is the electron gyromagnetic ratio).

The dipole magnetic field produced by the magnetic moment $\vec{m}$ is given by

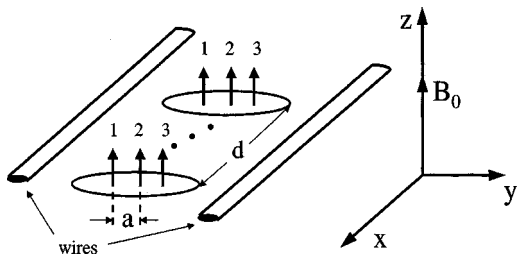

FIG. 1. A possible arrangement of electron magnetic moments in the external magnetic field, B_0. a is the distance between radicals in the three-spin group; d is the distance between the neighboring groups.

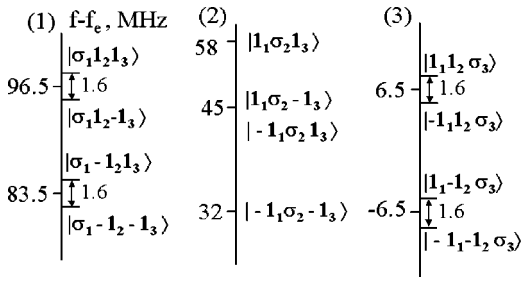

FIG. 2. Possible frequency differences, $(f-f_e)$, for three spins of a single group. The notation $(\sigma_1\sigma_2\sigma_3)$ indicates the orientations of three spins for the corresponding frequency.

$$\boldsymbol{B}=\frac{\mu_0}{4\pi}\frac{3\boldsymbol{n}(\boldsymbol{mn})-\boldsymbol{m}}{r^3}. \tag{1}$$

It is known that if the frequency difference between the spins is much greater than the dipole-dipole interaction, one can ignore the interaction between the transverse components of the spins (see, for example, Ref. 8).

For the dipole field produced by two neighboring spins in the same group we obtain

$$B_{z1}=(\mu_0/4\pi)(2\mu_B/a^3)(\sigma_2+\sigma_3/8),$$

$$B_{z2}=(\mu_0/4\pi)(2\mu_B/a^3)(\sigma_1+\sigma_3),$$

$$B_{z3}=(\mu_0/4\pi)(2\mu_B/a^3)(\sigma_1/8+\sigma_2). \tag{2}$$

Here B_{zk} is the dipole field at spin "k;" we count spins in Fig. 1 from the left to the right; $\sigma_k=-1$ if spin "k" is in its ground state ($|0_k\rangle$), and $\sigma_k=1$ if spin "k" is in its excited state ($|1_k\rangle$); $k=1,2,3$.

Next, we assume that the magnetic field gradient, $\partial B_z/\partial y=10^6$ T/m, $a=1.6$ nm, and $d=10$ nm. Suppose that the external magnetic field within each group increases from right to left in Fig. 1. Suppose also that the ESR frequency without the dipole–dipole interaction is f_e for the right (third) spin. The corresponding frequencies for the other two spins are approximately f_e+45 MHz and f_e+90 MHz. Taking into consideration the dipole fields in (2) we obtain the following approximate values for possible ESR frequencies of the three spins:

$$(1)\,f_e+90\pm 6.5\pm 0.8\ \text{MHz},$$

$$(2)\,f_e+45\pm 13\ \ \text{MHz},$$

$$(3)\,f_e\pm 6.5\pm 0.8\ \ \text{MHz}. \tag{3}$$

The allowed ESR frequencies for these three spins are shown in Fig. 2. The frequency shift due to the dipole field from two neighboring groups is negligible ($\sim$200 kHz). We will also assume that the ESR linewidth, δf, is about 1 MHz. The ESR linewidth can be caused by noise in the sample and fluctuations of the external magnetic field. The ESR linewidth of the order 1 MHz implies requirements on the fluctuations of the external magnetic field, $\Delta B<10^{-5}$ T. This is a challenge for experimental implementation of our proposal.

IV. CREATION AND DETECTION OF THE ENTANGLED STATE

Now we describe a protocol for creating an entangled state in all three-spin groups. We assume that all spins are initially in their ground state, $|000\rangle$. We start by applying a $\pi/2$-pulse to the first spin. The frequency of this pulse corresponds to the ground state of the neighboring spins ($f\approx f_e+83$ MHz, in our estimation). The Rabi frequency, Ω (i.e., the frequency of spin precession around the rotating transverse magnetic field in the rotating reference frame) must be greater than the ESR linewidth, but smaller than the frequency separation from the closest resonant frequency. We assume that $\Omega/2\pi=5$ MHz. Then, the duration of a $\pi/2$-pulse is $\pi/2\Omega=50$ ns. After this $\pi/2$-pulse, the state of the spins is described by the wave function,

$$\frac{1}{\sqrt{2}}(|0_1 0_2 0_3\rangle+i|1_1 0_2 0_3\rangle). \tag{4}$$

(Below we omit the index $k=1,2,3$ which indicates the position of the kth spin in the three spin state.) Next, we apply a π-pulse to the second spin. The frequency of this pulse is $f\approx f_e+45$ MHz. This π-pulse changes the state $|100\rangle$, so that the wave function becomes

$$\frac{1}{\sqrt{2}}(|000\rangle-|110\rangle). \tag{5}$$

Finally, we apply a π-pulse with the frequency $f\approx f_e+7$ MHz. This pulse acts on the state of the right spin if the neighboring spin is in the excited states. After this pulse, we obtain the completely entangled state,

$$\frac{1}{\sqrt{2}}(|000\rangle-i|111\rangle). \tag{6}$$

The separation between the frequency of the pulse and the closest resonant frequency is approximately 25, 13, and 14 MHz for the first, second, and third pulse, respectively. Thus, for all three pulses this separation is greater than the Rabi frequency, Ω. Note that the whole macroscopic spin system will consist of spin groups which are in identical entangled states.

To measure this entangled state one can use the fact that all components of the macroscopic magnetization are zero in this state. As an example, if we apply the same protocol replacing the second pulse (a π-pulse) by a 2π-pulse, the final state becomes an "ordinary" state,

$$\frac{1}{\sqrt{2}}(|000\rangle-i|100\rangle)=\frac{1}{\sqrt{2}}(|0\rangle-i|1\rangle)\otimes|0\rangle\otimes|0\rangle.$$

In this case, during the time $\sim 1/\pi\delta f$ after the pulse sequence one can observe the precession of the macroscopic magnetization of the left spins of the spin groups (if $\delta f\approx 1$ MHz, the time of the observation is ~ 0.3 μs). One can also observe the static z-component of magnetization produced by other two spins in all groups of the sample. If one

creates the entangled state (6), one cannot observe the transverse magnetization. The *z*-component of magnetization can only be observed after a time interval of the order of the spin longitudinal relaxation time.

V. SUMMARY

We discuss the opportunity for creation of an entangled spin state in SAM molecular systems. We consider SAM systems containing spin radicals. We take into consideration the magnetic dipole–dipole interaction between the spins of the same group, while neglecting the dipole interaction between the groups. A high magnetic field is applied to the system at low temperature, providing the initial ground state of the spins. The magnetic field gradient allows us to selectively excite any spin in a group. We show that a sequence of three resonant pulses can drive this system into an entangled state. We show how to measure this state. We believe that the experimental observation of three spin entanglement will be important step toward the application of long chains of electron or nuclear spins in SAM for quantum computation and quantum information processing. As an example, a proposal for a scalable solid-state quantum computer with paramagnetic atoms trapped by C_{60} molecules, suggested in Ref. 7, could also be implemented in SAM containing radicals instead of paramagnetic atoms.

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