Spin switching in a Mn-doped quantum dot using the optical Stark effect

D. E. Reiter,[1] T. Kuhn,[1] and V. M. Axt[2]

[1]*Institut für Festkörpertheorie, Universität Münster, Wilhelm-Klemm-Straße 10, D-48149 Münster, Germany*

[2]*Theoretische Physik III, Universität Bayreuth, D-95440 Bayreuth, Germany*

(Received 9 September 2011; revised manuscript received 20 December 2011; published 9 January 2012)

In a single quantum dot doped with a single Mn atom, ultrafast optical control can be used to coherently manipulate the Mn spin state. Here we show that due to the optical Stark effect induced by a detuned laser pulse the Mn spin can be efficiently flipped by one during the action of the pulse. We discuss the influence of different pulse envelopes on the flipping process. We then show how this flipping mechanism can be used in a switching protocol to address all Mn spin states selectively and point out the advantage of exploiting the optical Stark effect compared to previously proposed methods.

DOI: 10.1103/PhysRevB.85.045308 PACS number(s): 78.67.Hc, 75.50.Pp, 75.75.−c, 78.47.J−

I. INTRODUCTION

The fabrication of semiconductor quantum dots doped with a single Manganese (Mn) atom[1–3] has stimulated extensive research in the field of single spin control both experimentally[4–8] and theoretically.[9–15] After a detailed characterization of the states in a Mn-doped quantum dot,[1,2,16] recently the creation of a nonequilibrium occupation of the Mn spin states by incoherent relaxation during optical pumping has been demonstrated.[5,6]

Besides using resonant laser pulses to excite and deexcite excitonic states in a quantum dot, the application of detuned light fields has emerged as a powerful tool to manipulate the energetic structure via the optical Stark effect (OSE). Dressed excitonic states have been observed via spectral oscillations in two-color pump-probe signals from interface quantum dots[17] and via power-dependent Rabi splittings, Autler-Townes doublets, or Mollow triplets in neutral[18–22] and charged[23,24] self-assembled quantum dots. The OSE has been used to create entangled photon pairs from an anisotropic quantum dot[25] and recently an Autler-Townes splitting has also been observed in a single Mn-doped quantum dot.[8]

The ultrafast coherent control of a single spin is of great fundamental interest and one of the ultimate goals in the field of spintronics, in particular if spin states are used for the implementation of basic quantum information storage and processing. In previous works we have proposed a scheme to optically switch the Mn spin from a given eigenstate into all other Mn spin states[14,26] and showed that these dynamics can be monitored in time-resolved optical signals.[27,28] By ultrafast excitation of the quantum dot excitons exchange-induced Rabi oscillations describing a simultaneous flip of electron or hole spin and Mn spin are induced. At zero magnetic field the amplitude of the Rabi oscillations is typically rather small, because the exchange-coupled bright and dark states are strongly detuned.[29] This means that the Mn spin is essentially not flipping. However, by employing a series of laser pulses a complete flip of the Mn spin can be achieved.[14] The drawback is the very large number of pulses required in the case of large detuning. An alternative method to flip the Mn spin is to tune the exchange-coupled states into resonance by applying an external magnetic field.[14] In this case complete Rabi oscillations occur; however, the Mn spin does not remain in the desired final state but continues to oscillate until the resonance condition is removed by changing the magnetic field which, however, is typically not possible on ultrafast time scales.

An alternative way to bring the involved states into resonance is to use the OSE. Due to the OSE the energy of the bright states can be shifted during the action of a detuned strong pulse. Therefore in this paper we propose to use a detuned strong laser pulse to bring the exchange-coupled bright and dark exciton states temporarily into resonance opening up new opportunities for a controllable and precise switching of the Mn spin.

II. MODEL

Following our previous studies the quantum dot is modeled by one conduction band state with spin $S^e = \frac{1}{2}$; $S^e_z = \pm\frac{1}{2}$ and two valence band states, one of heavy-hole (HH, $S^h = \frac{3}{2}$; $S^h_z = \pm\frac{3}{2}$) and one of light-hole (LH, $S^h = \frac{3}{2}$; $S^h_z = \pm\frac{1}{2}$) type. Valence band mixing effects are neglected. The 15 states of the excitonic system are given by the ground state 0 without exciton, eight single-exciton states $H\pm1$, $H\pm2$, $L\pm1$, and $L\pm0$, and six biexciton states $HH0$, $LL0$, $HL\pm1$, and $HL\pm2$. All states are labeled by their valence band character and their total angular momentum. In the case of the $L\pm0$ state the sign refers to the hole spin. The Mn spin is $M = \frac{5}{2}$ and has six spin states $M_z = \pm\frac{5}{2},\pm\frac{3}{2},\pm\frac{1}{2}$. The full basis consists of product states $|X;M_z\rangle$ of the excitonic states $|X\rangle$ and the Mn spin states $|M_z\rangle$. The exchange interaction between the electron-hole spin and the Mn spin is given by

$$\begin{aligned} H_{e/h-\mathrm{Mn}} &= j_{e/h}\vec{S}^{e/h}\cdot\vec{M} \\ &= j_{e/h}\left[S^{e/h}_z M_z + \tfrac{1}{2}(S^{e/h}_+ M_- + S^{e/h}_- M_+)\right]. \end{aligned} \quad (1)$$

The Ising-type terms $\sim S^{e/h}_z M_z$ are diagonal in this basis and shift the energies. The flip-flop terms $\sim(S^{e/h}_+ M_- + S^{e/h}_- M_+)$ lead to simultaneous flips of the electron-hole spin and the Mn spin. We also account for the electron-hole exchange interaction $j_{eh}\vec{S}^e\cdot\vec{S}^h$. For details of the model and parameters we refer to Refs. 14 and 28. The light-field coupling is treated in the usual dipole and rotating wave approximation. By circularly polarized laser pulses, excitons with an angular momentum of ±1 can be excited, while the Mn spin does not couple to the light field. This model has been successfully used to explain measured photoluminescence spectra.[1,16]

We consider two types of pulses which we distinguish by the purpose of their action. For the purpose of exciton creation or destruction we use, as in previous studies, π pulses having a Gaussian shape with maximum amplitude E_π and a temporal width (full width at half maximum, FWHM) of $\tau_\pi = 100$ fs. The central laser frequency is set to the exciton energy of the bare single exciton E_X. In addition we will use laser pulses exploiting the OSE. Their central frequency is blueshifted with respect to the exciton energy E_X by 5 meV. The shape and the parameters of the OSE pulse will be specified below.

III. RESULTS

A. Rectangular pulse

A well-defined initial state for the dynamics that can be prepared experimentally is the one where the Mn spin takes one of its extremal values $M_z = \pm 5/2$, while no exciton is in the system. Here we choose as initial state $|0\rangle = |0; -\frac{5}{2}\rangle$. When the system is excited by a σ^--polarized π pulse to the bright HH exciton $|1\rangle = |H-1; -\frac{5}{2}\rangle$, the latter state couples to $|2\rangle = |H-2; -\frac{3}{2}\rangle$ via the flip-flop term of the electron-Mn exchange interaction. For zero magnetic field, because of the Ising terms of the exchange interactions, the two states $|1\rangle$ and $|2\rangle$ are energetically split by $\Delta E = 1.84$ meV. In the time domain off-resonant exchange-induced Rabi oscillations between these states build up. When now an OSE pulse is applied, the energy of state $|1\rangle$ is shifted due to the interaction with the off-resonant light field, such that it may come into resonance with state $|2\rangle$. Because the other states of the system influence the dynamics of this three-level system only marginally, we concentrate our discussion on these three states. A sketch of this effective three-level system is shown in Fig. 1.

We are interested in the dynamics of the system under the combined action of the resonant π pulse and a subsequent OSE pulse. In the simplest case the shape of the OSE pulse has a rectangular envelope with an amplitude E_{OSE} applied during a finite interval $t_1 \leqslant t \leqslant t_2$. As we will show below, resonance conditions between the states $|1\rangle$ and $|2\rangle$ are achieved for an amplitude $E_{\mathrm{OSE}}/E_\pi = 0.41$. Therefore, let us begin with such a pulse applied during the interval between $t_1 = 10$ ps and $t_2 = 25$ ps. The left column of Fig. 2 shows the resulting time-dependent occupations of the Mn spin states $M_z = -\frac{5}{2}$ and $-\frac{3}{2}$ and of the $H-1$ exciton as well as the used laser pulse sequence. At $t = 0$ a σ^--polarized π pulse excites the $H-1$ exciton and the exchange-induced Rabi oscillations set in. As the states $|1\rangle$ and $|2\rangle$ are off-resonant, the amplitude of the oscillations is very small with a value of about 0.01. At $t_1 = 10$ ps the OSE pulse is switched on. During the OSE

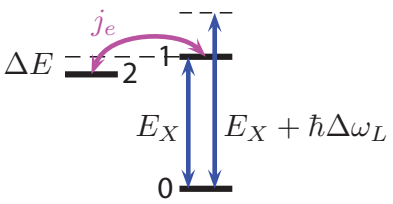

FIG. 1. (Color online) Scheme of the electronic states that dominate the switching process and their mutual couplings.

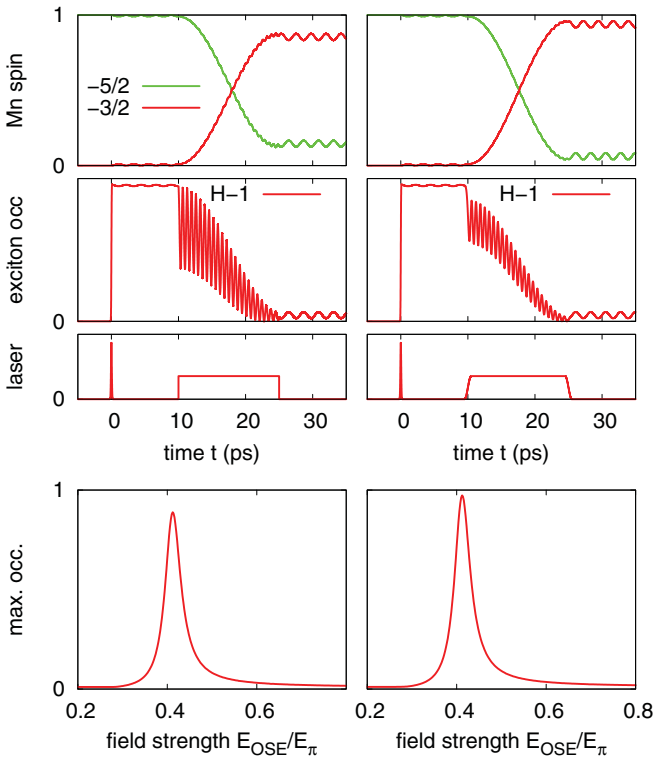

FIG. 2. (Color online) Upper panels: Occupations of the Mn spin states $M_z = -\frac{5}{2}$ and $-\frac{3}{2}$ and of the $H-1$ exciton state under the action of a π pulse and an OSE pulse. Lower panels: Maximal occupation of $M_z = -\frac{3}{2}$ as a function of the field strength E_{OSE}/E_π. Left: Rectangular OSE pulse. Right: Rectangular OSE pulse with softened edges.

pulse the states $|1\rangle$ and $|2\rangle$ are brought to resonance and thus the amplitude of the Rabi oscillations is close to 1. Indeed, during the pulse the occupation of $M_z = -\frac{3}{2}$ increases to 0.88 at $t = t_2 = 25$ ps, when the OSE pulse ends. After the pulse again an oscillation with a very small amplitude is seen as the exchange-coupled states are strongly off-resonant again. Thus the Mn spin has increased by almost one during the action of the OSE pulse. In Fig. 2 we additionally observe a fast oscillation on the occupation of the $H-1$ exciton. This oscillation is present also in the occupation of the ground state $|0\rangle$ (not shown) which clearly demonstrates that, though being off-resonant, the strong OSE pulse gives rise to pronounced off-resonant light-induced Rabi oscillations between the states $|0\rangle$ and $|1\rangle$.

The maximal occupation of the Mn spin state with $M_z = -\frac{3}{2}$ strongly depends on E_{OSE}, because the field strength determines the energy shift of the state $|1\rangle$. Only if the shift is such that the energies of the states $|1\rangle$ and $|2\rangle$ are close to resonance is an essentially complete switching possible. In the lower panel of Fig. 2 we have plotted the maximal occupation of the Mn spin state $M_z = -\frac{3}{2}$ achievable by a rectangular OSE pulse as a function of E_{OSE}. We can clearly identify the maximum for $E_{\mathrm{OSE}}/E_\pi = 0.41$, i.e., the value that has been taken above.

Sharp edges of a laser pulse often induce undesired effects due to the broad frequency spectrum associated with such an edge. Therefore, we now consider a rectangular pulse with

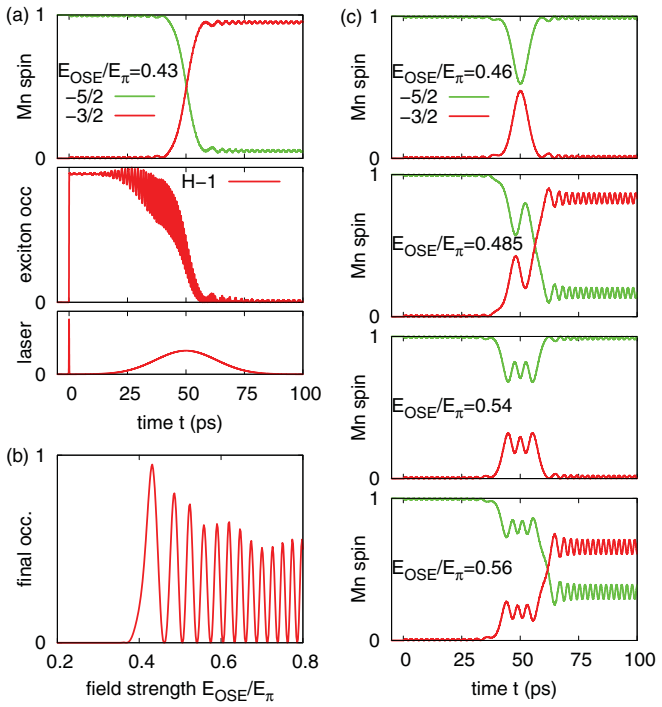

FIG. 3. (Color online) (a) Occupations of the Mn spin states $M_z = -\frac{5}{2}$ and $-\frac{3}{2}$ and of the $H-1$ exciton state under the action of a π pulse and a Gaussian OSE pulse. (b) Final occupation of $M_z = -\frac{3}{2}$ as a function of the field strength E_{OSE}/E_π. (c) Occupation of the Mn spin for different field strengths $E_{\mathrm{OSE}}/E_\pi = 0.46$, 0.485, 0.54, and 0.56.

softened edges which is also more realistic when compared to experimentally achievable rectangular pulses.[30,31] For this purpose we replace the steplike increase and decrease around t_i, $(i = 1,2)$ by $\frac{E_{\mathrm{OSE}}}{2}\{1 \pm \sin[\frac{\pi}{2b}(t - t_i)]\}$ during the intervals $|t - t_i| \leqslant b$. Here the upper sign refers to $i = 1$ and the lower one to $i = 2$. We take the same pulse parameters as before and set the roll-off time to $b = 0.5$ ps. In the right column of Fig. 2 we see that indeed the softening of the edges has a noticeable influence on the Mn spin dynamics. The oscillation between the states $|0\rangle$ and $|1\rangle$ is smaller and the final occupation of $M_z = -\frac{3}{2}$ after the pulse now reaches a maximum of 0.96.

B. Gaussian pulse

While nowadays pulses with an almost arbitrary shape can be formed, e.g., by using spatial light modulators,[30] and indeed ultrafast coherent control experiments using rectangular pulses have already been performed on quantum well structures,[32] typical experimental pulse shapes are usually closer to a Gaussian or a hyperbolic secans. Therefore we now consider a Gaussian pulse envelope with a temporal FWHM of 22 ps and a maximum field strength of $E_{\mathrm{OSE}}/E_\pi = 0.43$, the maximum being at $t = t_0 = 50$ ps. In Fig. 3(a) the corresponding occupations of the Mn spin states $M_z = -\frac{5}{2}$ and $-\frac{3}{2}$ and of the $H-1$ exciton state are shown. At $t = 0$ the π pulse again excites the $H-1$ exciton. Already around $t = 15$ ps a small oscillation of the occupation of $H-1$ sets in. A drastic increase of the Mn spin starts around $t = 40$ ps and ends around $t = 60$ ps, when $M_z = -\frac{3}{2}$ is almost fully occupied. During that interval the states $|1\rangle$ and $|2\rangle$ are close to resonance. After that the oscillation amplitude is again very small. The amplitude of the oscillation of the occupation of $H-1$ first increases due to the increasing pulse amplitude and then decreases as the exciton has changed to $H-2$. To analyze the efficiency of the spin switching using a Gaussian OSE pulse the final occupation of $M_z = -\frac{3}{2}$ after the pulse is shown in Fig. 3(b) as a function of the field strength. We find that the final occupation first increases monotonically, reaches a maximum at $E_{\mathrm{OSE}}/E_\pi = 0.43$, and then oscillates with increasing field strength. From Fig. 2 we know that the exchange-coupled states are in resonance at a field of $E_{\mathrm{OSE}}/E_\pi = 0.41$. Therefore, for field strengths above 0.41 the energetic positions of the states exhibit the following behavior: At the onset of the pulse the states $|1\rangle$ and $|2\rangle$ are off-resonant with $|1\rangle$ being above $|2\rangle$. At a field of $0.41E_\pi$ they become resonant. At the pulse maximum they are off-resonant again, but now the state $|1\rangle$ is below the state $|2\rangle$. When the Gaussian envelope decreases the two states become resonant for a second time and finally return to their initial off-resonant situation.

The influence of this behavior on the exchange-induced Rabi oscillations can be seen in the occupation of the Mn spin for different maximal field strengths $E_{\mathrm{OSE}}/E_\pi = 0.46$, 0.485, 0.54, and 0.56 shown in Fig. 3(c). At $E_{\mathrm{OSE}}/E_\pi = 0.46$ there is the first local minimum in the final occupation [see Fig. 3(b)]. When looking at the occupation of the Mn spin states $M_z = -\frac{5}{2}$ and $-\frac{3}{2}$ we see that during the pulse first the occupation of $M_z = -\frac{3}{2}$ increases at the expense of $M_z = -\frac{5}{2}$. At the pulse maximum at $t = 50$ ps both states reach an occupation of about 0.5. Then the occupations go back to their initial values; i.e., the occupation of $M_z = -\frac{5}{2}$ increases to almost 1 and the occupation $M_z = -\frac{3}{2}$ goes back to zero. This is because during the period when the energetic order of the states is inverted half a Rabi cycle is performed leading to the back rotation when the states are in resonance for the second time. For a maximal field strength $E_{\mathrm{OSE}}/E_\pi = 0.485$ the time where the states are energetically in resonance becomes shorter, while the time when the energetic order of the states is reversed becomes longer. Thus during the latter time interval the off-resonant Rabi oscillations perform a full rotation resulting in a high value of the final occupation of $M_z = -\frac{3}{2}$. Depending on the number of cycles of the exchange-induced Rabi oscillations between the resonance times, the final occupation of $M_z = -\frac{3}{2}$ can be close to 1 or close to 0. This is confirmed by looking at the dynamics at the third minimum and maximum of the final occupation at the maximal field strengths of $E_{\mathrm{OSE}}/E_\pi = 0.54$ and 0.56, respectively. In these cases we see that between the resonance times three Rabi cycles are performed. This explains why the final occupation oscillates as a function of the field strength and why also for very high field strengths a high final occupation is achievable.

C. Pulse sequence

In previous studies we have shown that by a certain switching scheme using pulse sequences on the HH and the LH exciton transitions the Mn spin can be switched from a given initial state into all other eigenstates.[14] Here, we want

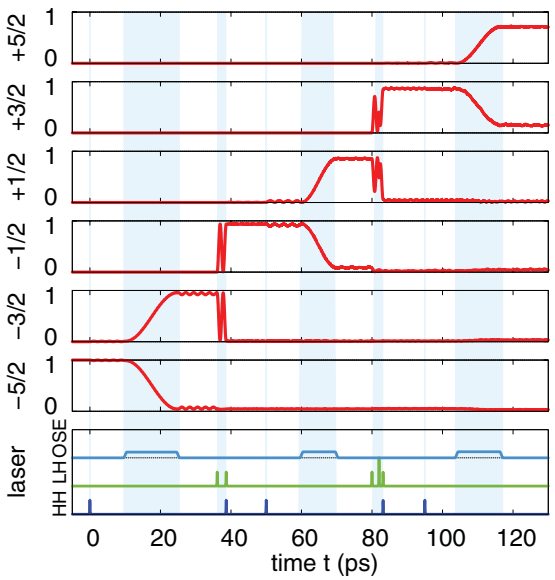

FIG. 4. (Color online) Occupation of the Mn spin states M_z under the laser pulse sequence shown in the lowest panel. Shaded intervals indicate times of the switching process, while during the white intervals the Mn spin is essentially constant.

to use the same switching scheme but now employ OSE pulses to control the exchange-induced Rabi oscillations. As we have seen above, different pulse shapes like rectangular or Gaussian pulses can be used for this purpose. The physics, however, is more transparent in the case of rectangular pulses. In addition, rectangular pulses have another advantage in our case. To clearly separate exciton generation and exchange-induced Rabi oscillation and thus to avoid a possible unwanted interference between these processes the π pulse and the OSE pulse should not overlap in time. Because of the long tails of Gaussian OSE pulses this requires rather large time delays between π pulse and OSE pulse. In the case of softened rectangular pulses the time delay is only limited by the rise time of the pulse which gives rise to considerably shorter total switching times. Therefore in the following we will employ sequences of Gaussian π pulses and softened rectangular OSE pulses.

In Fig. 4 the occupations of the Mn spin states are shown together with the employed laser pulse sequence, consisting now of π pulses on the HH exciton transition, π and 2π pulses on the LH exciton transition, and OSE pulses slightly above the HH exciton transition. We start with $M_z = -\frac{5}{2}$ and initialize the switching process with a π pulse on the HH transition at $t = 0$. By an OSE pulse acting between $t_1 = 10$ ps and $t_2 = 25$ ps with $E_{\mathrm{OSE}}/E_\pi = 0.41$ we switch from $M_z = -\frac{5}{2}$ to $M_z = -\frac{3}{2}$. In the next step additionally a LH exciton is excited at $t = 35$ ps by a π pulse on the LH transition leading to oscillations between $M_z = -\frac{3}{2}$ and $M_z = -\frac{1}{2}$. When $M_z = -\frac{1}{2}$ is occupied, the excitonic system is in the combined biexiton state $HL-2$, which can be deexcited to the excitonic ground state by two π pulses, one resonant on the HH and one on the LH transition. Subsequently we switch from $M_z = -\frac{1}{2}$ to $M_z = +\frac{1}{2}$ again exploiting a π pulse on the HH transition and an OSE pulse, here with a length of 10 ps and an amplitude of $E_{\mathrm{OSE}}/E_\pi = 0.46$. The different pulse parameters can be attributed to the different energy splitting between bright and dark exciton in this case. At $t = 70$ ps the Mn spin state $M_z = +\frac{1}{2}$ is fully occupied. Then we again use the LH flip to switch into $M_z = +\frac{3}{2}$. Here for the LH flip we use an additional 2π pulse to achieve a high occupation. Finally the last switch using an OSE pulse goes into $M_z = +\frac{5}{2}$, which is almost fully occupied from $t = 120$ ps on.

In Fig. 1 of Ref. 14 the same switching scheme has been shown using sequences of 2π pulses instead of the OSE pulses. There, in total 49 2π pulses on the HH exciton transition have been necessary to achieve the switching. In contrast, now only three OSE pulses are required. Furthermore, the total switching time is shorter. Note that we have included break times in the switching (white intervals in Fig. 4) when the Mn spin is essentially in an eigenstate, which can be made arbitrarily short, such that the total switching time is estimated without these times. Then the total switching time (shaded intervals) is about 45 ps, which is twice as fast as when switching with 2π pulses at zero magnetic field. Compared to the switching using a high magnetic field (Fig. 2 in Ref. 14), we find that here each Mn spin state can be addressed selectively with a reasonable accuracy. This is not possible for all states when using a magnetic field, because for some states a large back oscillation is inevitable.

IV. CONCLUSIONS

In summary, we have shown that a detuned laser pulse can be used to efficiently enhance the switching of the Mn spin due to the OSE. For a rectangular pulse an immediate shift of the exchange-coupled states into resonance increases the amplitude of the exchange-induced Rabi oscillations to almost one; i.e., the Mn spin is flipped by one. By softening the edges of the pulse the efficiency can be further increased. For a Gaussian pulse the Stark shift changes continuously, which also enabled almost complete flips of the Mn spin at various field strengths. When using the OSE in our recently proposed switching scheme, we find that a fast and selective addressing of each Mn spin state is possible.

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