Van der Waals (vdW) materials include a family of materials with various bandgap, and exhibit diverse electronic properties from semimetal (e.g., graphene) to semiconductor (e.g., transition metal dichalcogenides, TMDCs for short), and to insulator (e.g., hexagonal boron nitride, or hBN) [1]. Their common feature is the layered structure. These features make them widely applied in the fabrication of nano-photonic and electronic devices, particularly, vdW heterojunctions. HBN is the only layered material to date that is demonstrated to contain optically-detected electronic spins, which can benefit the construction of solid qubit and quantum sensor, etc., especially embedded in the nano-layered-devices. To realize this, Rabi oscillation is a crucial step. Here, we demonstrate the Rabi oscillation of $V_B^-$ spins in hBN. Interestingly, we find the behaviors of the spins are completely different under the conditions of weak and relatively higher magnetic field. The former behaves like a single wide peak, but the latter behaves like multiple narrower peaks (e.g., a clear beat in Ramsey fringes). We conjecture a strong coupling of the spin with the surrounding nuclear spins, and the magnetic field can control the nuclear spin bath.

Rabi oscillation of $V_B^-$ spin in hexagonal boron nitride

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VdW materials are a family of materials ranging from semimetal, semiconductor to insulator, and their common characteristic is the layered structure. These features make them widely applied in the fabrication of nano-photonic and electronic devices, particularly, vdW heterojunctions. HBN is the only layered material to date that is demonstrated to contain optically-detected electronic spins, which can benefit the construction of solid qubit and quantum sensor, etc., especially embedded in the nano-layered-devices. To realize this, Rabi oscillation is a crucial step. Here, we demonstrate the Rabi oscillation of $V_B^-$ spins in hBN. Interestingly, we find the behaviors of the spins are completely different under the conditions of weak and relatively higher magnetic field. The former behaves like a single wide peak, but the latter behaves like multiple narrower peaks (e.g., a clear beat in Ramsey fringes). We conjecture a strong coupling of the spin with the surrounding nuclear spins, and the magnetic field can control the nuclear spin bath.

Among this family of layered materials, hBN has a large bandgap of ~6 eV, which makes it have the ability to host plenty of kinds of defects, just similar to diamond [29, 31] and silicon carbide [32, 34], etc. Single-layer hBN was first found to emit single photons at room temperature in 2016 by Tran et al. [29], and after that, a great of interest was stimulated to treat hBN defects as the promising single-photon emitters [16, 25], and furthermore, as the potential solid spin qubit [35, 43]. As the single-photon source, hBN defect (in monolayer, flake or bulk) has the advantages of high brightness [21, 24], broad spectral range [20], easy tunability [21, 22], and easy fabrication, etc. The fabrication methods are diverse, including chemical etch [15], electron or ion irradiation [18, 19, 25, 40], laser ablation [19, 21], strain [29], and so on.

Especially, defects in hBN attract a lot of attentions to be a good candidate for solid spin qubit (particularly, in the vdW-nano-devices). Actually, electron paramagnetic resonance (EPR) signals in hBN have been found in very early decades [44–46]. These signals are recognized by numerical calculations recently [47, 48], and these theoretical works also predict many possible defects in hBN who have the potential to give optically detected magnetic resonance (ODMR) signals. In experiment, Exarhos et al. [35] find the magnetic-field-dependent intensity of fluorescence emitted from a hBN defect in 2019. Later, the ODMR signals are revealed by Gottscholl et al. [36, 37], Chejanovsky et al. [38] and Mendelson et al. [39], respectively. The first kind of defects are assumed to be $V_B^-$, and the third kind of defects are conjectured to be related to carbon. Based on the experimental results, several theoretical analyses are carried out, especially for $V_B^-$ [39, 50], and the temperature-dependent features of this kind of spin defect are also detailedly investigated recently in experiment [42].

The energy levels of $V_B^-$ is gradually clear, and a simplified diagram is sketched in Fig. 1(a). As discussed in Refs. [37, 49], this defect contains a triplet ground state (GS), which is primarily constituted of three energy levels with $m_s = 0$ and $m_s = \pm 1$, and $D$ is the zero-field splitting (ZFS) between them. ES and MS represent the excited states and metastable states, respectively. The green arrows stand for the excitation laser which pump the population to above ES, and the red arrow repre-
FIG. 1: Simplified energy levels and ODMR results. (a) Simplified energy levels of $V_{\text{B}}^-$ center and the related optical transitions among ground states (GS), excited states (ES) and metastable states (MS). The 532-nm laser (green) is used for the spin polarization and readout, and the microwave (pink) is used for coherent control of the spin state. (b) ODMR spectrum measured at room temperature with no external magnetic field. The bimodal signal due to the local strain has obvious hyperfine structures, which indicate the nucleus-electron interaction between $V_{\text{B}}^-$ and the three neighboring nitrogen nuclei ($^{14}\text{N}$). The experimental data are fitted using two-Lorentzian function to obtain $\nu_1 \sim 3.424 \text{ GHz}$ and $\nu_2 \sim 3.533 \text{ GHz}$. (c) Dependence of the frequency shift of the $m_s = -1 \leftrightarrow m_s = 0$ transition on the magnetic field parallel to the hexagonal $c$ axis, from which we can fit the $g$ factor of $V_{\text{B}}^-$ to be $1.992 \pm 0.010$.

sent the fluorescence to be detected. The gray wavy arrows represent the inter-system crossings (ISC) between $S = 3$ and $S = 1$. The pink circled arrow is the applied microwave (MW) between $m_s = 0$ and $m_s = \pm 1$, which will change populations of these states, and hence change the intensity of fluorescence. By recording the difference of the fluorescence intensities, we can read the state of spin qubit. This method is called ODMR. A typical ODMR signal of $V_{\text{B}}^-$ at zero-magnetic-field is shown in Fig. 1(b). The excitation laser is always on, and the MW works at the on/off mode, then the contrast is calculated from the difference between the on and off fluorescence. $\nu_0 = (\nu_1 + \nu_2)/2 = 3.479 \text{ GHz}$ is the frequency corresponding to ZFS $D$, and $\nu_1, \nu_2$ correspond to the transitions between $m_s = \pm 1$ states to $m_s = 0$, respectively. Remarkably, for each transition peak, we can clearly see several hyperfine splittings, which can be identified as the nucleus-electron interaction. For $V_{\text{B}}^-$ defect, there are three nitrogen nuclei ($^{14}\text{N}$) around, each of which has a nuclear spin of $I = 1$. Therefore, totally $2(3I) + 1 = 7$ hyperfine transitions should be observed, and the interval between them is detected [37] and calculated [39] to be approximately 47 MHz. Fig. 1(c) shows the frequency shift of the $m_s = -1 \leftrightarrow m_s = 0$ transition as the magnetic field varies, from which we can fit the $g$ factor of this spin to be $1.992 \pm 0.010$.

The next step is naturally to coherently operate the solid spin, of which the Rabi oscillation is a key tool. Here we utilize the two-level states $m_s = -1, 0$ as the spin qubit to perform the coherent control. Fig. 2(a) is the time diagram of Rabi oscillation. After a long excitation laser pulse, the spin is polarized to $m_s = 0$ state, then a MW pulse with length of $\tau$ is applied to rotate the spin, followed by a readout pulse. The results are shown in Fig. 2(b). At magnetic field of $B = 0 \text{ mT}$, we see a standard decayed Rabi oscillation, but we also observe a tiny decay of background, which may be caused by the overlarge density of $V_{\text{B}}^-$ defects, since our integrated dose of neutron irradiation is quite large ($\sim 2.0 \times 10^{17} \text{ n cm}^{-2}$). For results of non-zero magnetic field, we observe an oscillation of multiple Rabi frequencies. We conjecture that a more orderly ensemble of defects is formed because of the magnetic field, which makes the hyperfine peaks narrower and can to some extent be separated from each
Microwave B 3(a) and Fig. 3(b) exhibits the relaxation result at our Rabi results. We find \( T_\mu T_m T \). By fitting this result, we derive relaxation time \( T \) pulse. Utilizing \( \pi \) on the \( m \) the microwave pulse with length \( \tau \).

FIG. 2: Rabi oscillations. (a) Pulse sequence of Rabi measurement comprising the first laser pulse for spin polarization, then the microwave pulse with length \( \tau \) for manipulation and the second laser pulse for state readout. (b) Rabi oscillations on the \( m_s = -1 \leftrightarrow m_s = 0 \) transition observed at room temperature and different magnetic field. The data are fitted using \( \Sigma_{n=1}^N A_n \exp(-\tau/T_1) \cos(2\pi f_1 \tau + \phi_1) + b \exp(-\tau/T_2) + c \), with \( n = 1, 2 \) and 3 (red lines) from top to down, respectively. \( A_n, T_1, f_1, \phi_1, b, T_2 \) and \( c \) are the amplitude, oscillation decay time, frequency, phase, decayed background and its decay time, constant background, respectively. (c) Rabi frequency measured with a linear dependence on the square root of microwave power \( \sqrt{P} \).

other; or maybe some other reasons such as the influences of the environmental nuclear or electronic spins [30]. By varying the MW power, we derive the linear dependence of the fitted Rabi frequency versus the square root of power \( \sqrt{P} \) (see Fig. 2(c)), which indicates the validity of our Rabi results.

With the Rabi frequency, we can define \( \frac{\pi}{2} \)-pulse and \( \pi \)-pulse. Utilizing \( \pi \)-pulse, we can measure the spin-lattice relaxation time \( T_1 \). The pulse sequence is shown in Fig. 3(a) and Fig. 3(b) exhibits the relaxation result at \( B = 0 \) mT. By fitting this result, we derive \( T_1 = 16.377 \pm 0.416 \) \( \mu s \). Then we repeat this sequence for various magnetic fields and obtain the results in Fig. 3(c). We find \( T_1 \) is approximately independent of the magnetic field. Next, we perform the sequence \( \frac{\pi}{2} - \frac{\pi}{2} - \pi - \frac{\pi}{2} - \frac{\pi}{2} \) (spin echo, see Fig. 3(d)) to measure \( T_2 \). At \( B = 0 \) mT, we observe a monotonic decay of contrast shown in Fig. 3(e), and \( T_2 \) is fitted to be \( 82.121 \pm 2.462 \) ns, which is quite short. We conjecture that it may also be caused by the overlarge density of \( V_B \) defects. At \( B = 36 \) mT, we find the decayed-contrast curve is complicatedly modulated (see Fig. 3(f)). We cannot fit it well, and the red line is only a guide for eyes. Here we want to note that, since \( T_2 \) is quite short, the impact of time durations of the MW pulses, especially the \( \pi \)-pulse, can not be ignored, therefore, the values of the fitted \( T_2 \) will have inaccuracy, but the order of magnitude can be determined. For \( T_1 \) which is far longer than MW-pulse durations, this problem is not met.

We also perform the Ramsey interference experiment on the \( V_B \) spins. The pulse sequence is presented in Fig. 4(a), and Fig. 4(b) shows the result at \( B = 0 \) mT. We have not seen the oscillations, which may be caused by the fast decay corresponding to a short \( T_2^* \). On one hand, the nucleus-electron interaction splits the \( m_s = -1 \leftrightarrow m_s = 0 \) transition into 7 peaks; on the other hand, every peak is broad at zero magnetic field due to the inhomogeneous ensemble, and the peaks adhere to each other to form a single wide peak and thus induce a short \( T_2^* \). Similar to the result in Fig. 2(b), we also see a slow decay of background in this figure (this data cannot be fitted well using single-decay curve, but a double-decay curve). The reason may also be attributed to the overlarge density of \( V_B \) defects in the sample. In contrast, when we apply a magnetic field of \( B = 44 \) mT and set the MW frequency at \( f_{MW} = 2200 \) MHz, we see a multiple-frequency oscillation, in which a beat is clearly recognized, and it is superposed on another slow oscillation. These three frequencies are fitted as \( f_{-1} = -44.171 \pm 0.039 \) MHz, \( f_0 = 0.934 \pm 0.131 \) MHz.
FIG. 3: $T_1$ measurement and spin echo detections. (a) Pulse sequence for characterizing the spin-lattice relaxation dynamics, including the spin polarization and readout, the microwave $\pi$-pulse obtained from Rabi-measurement and the free evolution time $\tau$ for changes. (b) $T_1$ measurement at 0 mT revealing the spin-lattice relaxation time of $T_1 = 16.377 \pm 0.416 \mu$s. (c) $T_1$ time versus magnetic field, suggesting that there is roughly no $T_1$ dependence on magnetic field. (d) Pulse scheme for spin echo measurement with $\pi/2 - \tau - \pi - \tau - \pi/2$ sequence, where $\tau$ is the free evolution time. (e) Optically-detected spin-echo measurement at 0 mT. (f) Spin echo at 36 mT which cannot be fitted well, showing complicated oscillations induced by the nuclear spin bath and the red line is only a guide for eyes.

MHz, $f_1 = 45.872 \pm 0.063$ MHz, respectively. We therefore conjecture that the three peaks that contribute to the Ramsey oscillation are located at $h(f_{MW} + (-)f_i)$ ($i = -1, 0, 1$, $h$ is Planck constant, and the sign “-” represents the possibility of reverse direction), and other peaks contribute to this oscillation little since they are much farther from the MW frequency. The distance between the adjacent peaks is $f_0 - f_{-1} = 45.105 \pm 0.136$ MHz$\approx f_1 - f_0 = 44.938 \pm 0.145$ MHz (the Planck constant is omitted here), which is coincident with the calculated [49] and observed [37] energy separation of the hyperfine splittings induced by nucleus-electron interaction (approximately 47 MHz). The fitted $T_2^*$ of these three peaks are $0.665 \pm 0.108$ $\mu$s, $2.500 \pm 2.160$ $\mu$s and $1.448 \pm 0.841$ $\mu$s, respectively. Although we have not extracted $T_2$ in the case that magnetic field is nonzero due to the quite complex modulations, we can conjecture that $T_2$ will be in the order of microseconds from the results of $T_2^*$ of the individual peaks. It seems the magnetic field help elongate the spin coherence time.

It is interesting to find that in both the Rabi- and Ramsey-oscillation results, the observed phenomena are very different when the magnetic field is weak or relatively strong. At zero magnetic field, the defects behave as a large ensemble with a single broad peak; but when the magnetic field is strong enough, the defects behave like a more orderly ensemble with multiple narrow peaks, i.e., the multiple-frequency fittings of each data with higher magnetic field. This phenomena suggest that the $V_B^-$ spin is highly correlated to the neighboring nuclear spins, which can be utilized to study the nuclear spins or the correlations between them.
FIG. 4: Ramsey interference. (a) Ramsey pulse sequence with $\pi/2 - \tau - \pi/2$. (b) Ramsey result with $B = 0 \text{ mT}$ and $f_{\text{MW}} = 3428 \text{ MHz}$. No oscillation is observed but a fast decay with $T_2^* = 60.198 \pm 2.747 \text{ ns}$. A slow background decay is also observed as that in Rabi results. (c) Ramsey result at 44 mT and 2200-MHz microwave frequency. Three frequencies are observed and two of them form a clear beat. The red line is the fitting. The distances between the adjacent frequencies are both around 47 MHz (the hyperfine splitting due to the nucleus-electron interaction observed and calculated previously). $T_2^*$ corresponding to these three frequencies are $0.665 \pm 0.108 \mu$s, $2.500 \pm 2.160 \mu$s and $1.448 \pm 0.841 \mu$s, respectively.

As the only candidate for spin qubit in vdW material (to date), the coherent operations of defects in hBN based on Rabi oscillation play the crucial role, and provide a powerful tool for the design and construction of spin-based vdW-nano-devices, especially when the techniques of vdW heterojunction are combined. Although $T_2$ is still quite short, which may be primarily due to the overlarge density of $V^{-}_B$ defects in the sample caused by the high-dose neutron irradiation, there will be several methods to improve it. For example, we can decrease the integrated dose of neutron irradiation; or perform a suitable annealing on the sample since high-temperature condition can reduce the $V^{-}_B$ defect number; or put the sample into a low-temperature cryostat; or apply higher magnetic field; etc.

In summary, we have realized the Rabi oscillation of the $V^{-}_B$ spins in hBN, based on which we also detect $T_1$ and perform the spin-echo and Ramsey-interference experiments. We find $T_1$ is almost not affected by magnetic field, which is roughly around $16.377 \pm 0.416 \mu$s; however, the results of Rabi oscillation, spin echo and Ramsey oscillation are very different under the conditions of weak and relatively strong magnetic field. At zero magnetic field, the defects behave as a large ensemble with a single broad peak, i.e., the Rabi result is well fitted by a single-frequency oscillation, the echo result shows a fast decay of $82.121 \pm 2.462 \text{ ns}$, and the Ramsey result also decays fast without any oscillations. When the magnetic field goes higher, the defects behave like a more orderly ensemble with multiple narrow peaks, and we see multiple-frequency oscillations in both Rabi and Ramsey results. Especially, we see a clear beat and an additional slow oscillation in the Ramsey result, and by fitting this result, we find the distances among these three frequencies ($45.105 \pm 0.136 \text{ MHz} \approx 44.938 \pm 0.145 \text{ MHz}$) are roughly equal to the energy separation of the nucleus-electron-interaction-induced hyperfine splitting (approximately 47 MHz, $h$ is omitted). The decay times ($T_2^*$) of these three oscillations are $0.665 \pm 0.108 \mu$s, $2.500 \pm 2.160 \mu$s and $1.448 \pm 0.841 \mu$s, respectively, from which we conjecture
that $T_2$ for each separated peak will be in the order of microsecond. It seems the magnetic field freezes the environmental spins to some extend and elongates the $T_2$. Our results suggest that the $g_B$ spin is highly correlated to the neighboring nuclear spins, which provides a potential tool to study them.

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