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David A. Broadway, Scott E. Lillie, Nikolai Dontschuk, Alastair Stacey, Liam T. Hall, Jean-Philippe Tetienne, and Lloyd C. L. Hollenberg

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David A. Broadway,1,2 Scott E. Lillie,1,2 Nikolai Dontschuk,1,2 Alastair Stacey,1,3 Liam T. Hall,2 Jean-Philippe Tetienne,2,a) and Lloyd C. L. Hollenberg1,2

1Centre for Quantum Computation and Communication Technology, School of Physics, University of Melbourne, Parkville, VIC 3010, Australia
2School of Physics, University of Melbourne, Parkville, VIC 3010, Australia
3Melbourne Centre for Nanofabrication, Clayton, VIC 3168, Australia

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Precise control of the resonant frequency of a spin qubit is of fundamental importance to quantum sensing protocols. We demonstrate a control technique on a single nitrogen-vacancy (NV) centre in diamond where the applied magnetic field is modified by fine-tuning a permanent magnet’s magnetisation via temperature control. Through this control mechanism, nanoscale cross-relaxation spectroscopy of both electron and nuclear spins in the vicinity of the NV centre is performed. We then show that through maintaining the magnet at a constant temperature, an order of magnitude improvement in the stability of the NV qubit frequency can be achieved. This improved stability is tested in the polarisation of a small ensemble of nearby 13C spins via resonant cross-relaxation, and the lifetime of this polarisation explored. The effectiveness and relative simplicity of this technique may find use in the realisation of portable spectroscopy and/or hyperpolarisation systems.

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Progress in quantum information processing (QIP) and quantum sensing (QS) has driven interest in spin qubits,1–4 which are attractive candidates to both of these endeavours. One such qubit is the nitrogen-vacancy (NV) centre in diamond,5 which has long coherence times at room temperature and has demonstrated the capability of measuring a range of nanoscale field values from magnetic fields,6–9 electric fields,10 temperatures,11 and can even achieve sub-mHz spectral resolution in nuclear magnetic resonance (NMR).12–14 Application of the NV centre in QIP and QS requires control of the spin sublevels of the system via the Zeeman effect,9 with magnetic fields between 0 and 0.4 T being typically applied depending on the application.15 This control is usually achieved using a permanent magnet, because electromagnets in this regime are generally not compatible with the geometrical constraints imposed by typical confocal microscopes employed to address single NV centres. However, the use of permanent magnets poses two problems. First, in experiments that require scanning of the B field [e.g., cross-relaxation (CR) spectroscopy15–19], the magnet must be physically moved which limits the precision of the field strength selection. Second, the magnetisation of permanent magnets is temperature dependent. Without adequate control, variations in ambient temperature will affect the stability of the qubit frequency especially over long measurement times.20 This instability can limit the resolution in nanoscale NMR spectroscopy21,22 and the efficiency of hyperpolarisation of external spins based on cross-relaxation.19

In this work, we use the temperature dependence of the field strength in a permanent magnet as a way to control the magnetic field in single NV experiments. We first demonstrate that active control of the magnet temperature allows the B field to be scanned in a range sufficient for CR spectroscopy, as we illustrate by recording CR spectra of both electron and nuclear spins. Next, we consider the long term stability of the NV frequency from room temperature variations and show that the resulting fluctuations can be mitigated via this active temperature control. Finally, we take advantage of this newly obtained long term stability to investigate the polarisation and depolarisation dynamics of the 13C nuclear spin bath via cross relaxation induced polarisation (CRIP), which garners valuable information about the evolution of polarised spin baths during NV dark periods.

The frequency of the NV electron spin transition |0⟩ → |−1⟩ (i.e., the qubit frequency) typically used in experiments is given by ωNV = D − γνB [Fig. 1(a)], which has a zero field splitting of D = 2.87 GHz with further splitting induced by a magnetic field along the quantization axis (B), with a defect gyromagnetic ratio γν. Temperature control of the magnetisation of a permanent magnet is achieved by attaching a thermoelectric cooler (TEC) to the magnet as shown in Fig. 1(b). The control of the temperature was implemented through a feedback from a thermistor attached to the magnet on the opposite side to the TEC. This feedback was passed to a controller (TE technology TC-720) for proportional integral derivative (PID) control with Ziegler-Nichols method tuning. The magnet is a 50 mm × 12.5 mm grade 3N8 neodymium (NdFeB) disc with a NiCuNi coating (AMF Magnetics). The specified temperature coefficient of the magnetisation Mₜ around room temperature is χ₀ = dM/dT = −0.12±0.1%/°C with a Curie temperature of 310–370°C.

To test the ability to control the magnet temperature, we apply a 1°C step function about ambient conditions [Fig. 1(c), red dashed line] using the PID controller and measure the thermal settling time of the magnet. Given the physical dimensions of the magnet, the thermal settling time for this modest temperature change (blue) is considerable (several minutes). For smaller temperature changes, in the regime <0.2°C, the settling time reduces to <1 min. The state dependent photoluminescence (PL) of the NV spin sublevels allows the transition frequency between them to be measured by optically detected

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a)Electronic mail: jtetienne@unimelb.edu.au
magnetic resonance (ODMR).9 Performing an ODMR sweep every 10 s during the temperature change shows a direct correlation between the NV frequency $\omega_{NV}$ and the measured temperature of the permanent magnet, see Fig. 1(d). Here, the 1°C change in temperature translates into a change of 2.5 MHz in $\omega_{NV}$ at a field of $B \sim 1020$ G (i.e., $\gamma_{NV} B \sim 2860$ MHz). We then mapped the response of the NV frequency to a ramp in temperature [Fig. 1(e)], showing a linear response that is quantitatively consistent with the magnet specifications [Fig. 1(f)]. This response demonstrates a magnetic field scanning range of 0.5% via varying the magnet temperature by only 4°C (from 22 to 26°C), which corresponds to a 10 MHz range at $B \sim 1020$ G [Fig. 1(e)]. With temperature variations up to 100°C achievable with standard TECs, ranges as large as 12.5% [i.e., $\sim$120 G in the magnetic range used in Fig. 1(e)] may be achieved, which is sufficient to detect the spectra of electron-nuclear coupled systems with a large hyperfine splitting.16 On the other hand, the smallest temperature step size achievable with our current setup is 0.01°C, corresponding to a change of 26 kHz. This is a twofold improvement over the resolution achieved in previous works using motorised translation stages,16,20 which are capable of a 400-nm axial step size or 65 kHz around the ground state level anti-crossing (GSLAC).23

We now demonstrate the use of temperature enabled magnetic field scanning to perform CR spectroscopy on a single NV spin.24 CR spectroscopy is a technique that monitors the relaxation time ($T_1$) of the NV spin as a function of $B$. This produces a spectrum of the surrounding spins, which are seen as decreases in the NV $T_1$ at each resonance $\omega_{NV} = \omega_{\text{env}}$, where $\omega_{\text{env}}$ is the Larmor frequency of an environmental spin. We first demonstrate the technique targeting on electronic spins located on the surface of the diamond [Fig. 2(a), red]. To this end, we use a near-surface NV centre ($\sim$5–10 nm deep17) and a permanent magnet fixed such that $B \sim 512$ G. The magnet temperature is then controlled to finely scan $B$ across the free electron resonance $\omega_{NV} = \gamma_{NV} B = 1435$ MHz. Figure 2(b) shows the spectrum obtained by measuring the PL intensity $I_{NV}$ following a free evolution time $t = 100 \mu$s, which maps changes in the $T_1$ time as $I_{NV} \propto \exp(-t/T_1)$. The dip centred at a temperature $T = 24^\circ$C (corresponding to $\omega_{NV} = 1435$ MHz as measured by ODMR) is the signature of unpaired electron spins assumed to be associated with surface termination states or dangling bonds. By tuning the magnetic field close to the GSLAC (where $\omega_{NV} < 5$ MHz), it is also possible to detect nuclear spin species [Fig. 2(a), black]. Figure 2(c) shows an ODMR map obtained from a deep NV centre ($\sim$1 μm from the surface) by scanning the temperature from 21 to 27°C near $B = 1024$ G. The avoided crossing in the higher temperature range (i.e., lower magnetic field) is characteristic of the GSLAC structure for the $^{14}$NV centre.23

FIG. 1. (a) Depiction of a nitrogen-vacancy (NV) centre in diamond excited with a green laser and subject to the field produced by a temperature controlled magnet. The NV state is read out via the red photoluminescence (PL). The NV $|\pm 1\rangle$ states are split by the magnetic field applied along the quantization axis, where the magnet produces less field at hotter temperatures. (b) Picture of the magnet attached to a TEC module. (c) Step function in the set temperature (red dashed line) and the corresponding thermistor temperature reading (blue). (d) Corresponding optically detected magnetic resonance (ODMR) map from a single NV centre during the temperature pulse showing the $|0\rangle \rightarrow |\pm 1\rangle$ spin transition with a background field $B \sim 1020$ G. (e) ODMR mapped as a function of the set temperature. (f) Black curve: relative change in magnetic field $B$ vs temperature, as extracted from (e) using the relationship $\omega_{NV} = D - \gamma_{NV} B$, giving a temperature coefficient of $\omega_{NV} = -0.094/%/C$. Red curve: relative change in magnetisation $M_s$ vs temperature, as calculated from the specified coefficient of our magnet ($\omega_{NV} = -0.12 \pm 0.1%/C$). Both plots are normalised by their respective values at $T = 22^\circ$C.

FIG. 2. (a) Depiction of the NV sensing external electrons (red) and internal $^{13}$C spins (dark grey). (b) CR spectrum ($t = 100 \mu$s) across the frequency range $\omega_{NV} = 1426–1445$ MHz (via temperature scanning between 18 and 32°C), showing a peak corresponding to unpaired electrons ($g = 2$). (c) $^{13}$NV hyperfine structure across the GSLAC as mapped by temperature scanned ODMR. (d) CR spectrum ($t = 2 \mu$s) across the GSLAC ($\omega_{NV}$ varied from −2 to 2 MHz), where the two outer peaks correspond to $^{13}$C spins. In (d), the measurement sequence alternates initialisation of the NV in $|0\rangle$ and $|\pm 1\rangle$ before the free evolution time [see Fig. 4(a–i)] to prevent net polari-sation transfer to the $^{13}$C bath.19 The data shown are the readout following initialisation in $|0\rangle$ only.
The CR spectrum, measured using $\tau = 2 \mu s$, reveals three features [Fig. 2(d)]. The central feature is caused by state mixing at the GSLAC and the two outer peaks are the signature of $^{13}$C nuclear spins in the diamond lattice, which come into resonance with the NV before and after the GSLAC.\textsuperscript{19,23}

A number of applications, such as high precision measurements,\textsuperscript{12} long decoupling sequences, and hyperpolarisation of external spins,\textsuperscript{19,25} require that the NV spin remains on resonance (with a driving field or a given spin species) for hours to days. While it is possible to post-process to remove off-resonance data, this is an undesirable solution for hyperpolarisation where off-resonant conditions limit the degree of polarisation transferred to the spin bath. By carefully controlling the temperature of the magnet, it is possible to significantly improve the long term stability of the NV frequency. In the absence of active temperature control, we observe variations in the room temperature [Fig. 3(a)] which result in changes in the NV frequency on the order of 2 MHz over the course of 20 h [Fig. 3(b), orange line]. Including the PID loop to maintain a constant temperature of the magnet (as measured by the thermistor) reduces the change in frequency to 800 kHz peak to peak [Fig. 3(b), green line]; however, there is still an effect on the magnetisation from changes in the room temperature (see supplementary material for details). This effect can be reduced by setting up a second order correction loop based on the measured ambient temperature, such that the changes to the magnet temperature set point are defined as

$$\Delta T_M = -\frac{\Delta T_R}{C_0} + \frac{\partial T_R}{\partial T},$$

where $\Delta T_M = 2.7$ MHz/$^\circ$C is the direct change in magnetisation from temperature, $T_M$ and $T_R$ are the thermistor reading of the magnet and room temperature, respectively, and $\Delta$ denotes difference from the original value. $\Delta T_R$ and $\partial T_R/\partial T$ give the change in magnetisation from the difference and derivative of the room temperature, respectively. This is simply a proportional derivative (PD) feedback loop acting on the set point, which is combined with the PID loop acting on the TEC. Using this second order feedback reduces the changes to 200 kHz [Fig. 3(b), blue line], offering an improvement in stability by an order of magnitude compared to a non-controlled temperature. This is a similar improvement to previous results using a temperature controlled stage with extensive insulation.\textsuperscript{26} Importantly, the remaining fluctuations in the NV frequency are within the intrinsic ODMR linewidth for typical near-surface NV centres ($\sim 500$ kHz), reducing the noise limit for long measurements to the intrinsic level. These fluctuations could be further reduced by simply changing to a common samarium-cobalt (SmCo) magnet which has a lower temperature coefficient $\chi_0 = 0.04\%/^\circ$C. This would return a stability of $\sim 60$ kHz and a resolution of $\sim 8$ kHz, which is close to the smallest linewidths achievable.\textsuperscript{27}

As a final experiment, we consider the polarisation of a small ensemble of $^{13}$C spins using cross relaxation induced polarisation (CRIP).\textsuperscript{19} By repeatedly initialising the NV in the same spin state and tuning its frequency to the $^{13}$C resonance, it is possible to gradually polarise the $^{13}$C bath, as demonstrated in Refs. 19 and 28. Since the polarisation buildup occurs over long time scales (typically hours), instabilities in the NV frequency often limit the maximum level of polarisation achievable and the ability to monitor its evolution under a variety of conditions. Here, we use the improved stability to investigate the lifetime of the $^{13}$C polarisation. We first characterise the NV-bath interaction by using a sequence in which the NV spin is initialised in |0\rangle and \{-1\} in alternance before the free evolution time $\tau$ [Fig. 4(a-i)]. By tuning the NV frequency on resonance with $^{13}$C [after the GSLAC, see Fig. 3(d)] and varying $\tau$, we observe a coherent oscillation [Fig. 4(b)] whose frequency denotes the total coupling strength to the $^{13}$C bath.\textsuperscript{19} Using an optimal evolution time $\tau_{opt} = 1.9\mu s$ (half the period of the flip-flop oscillation), we then apply $N$ polarisation steps to polarise the bath in the |↑⟩ state (by initialising the NV in |0⟩) followed by $N$ steps to polarise the bath in |↓⟩ (by initialising the NV in |−1⟩), see sequence in Fig. 4(a-ii). The resulting polarisation curve is plotted as a function of step number in Fig. 4(c) and shows saturation of the polarisation after about ten steps. Here, only the spins within the coupling strength of $\Gamma = 1/\tau_{opt}$ are probed, corresponding to a distance from the NV of $< 1$ nm. We also note that each polarisation step results in a maximum transfer of one quantum of polarisation, and thus with $N = 10$, there is a maximum of ten nuclear spins polarised.

The contrast between the polarised and un polarised spin bath states allows measurements of the lifetime of the polarisation. To achieve this, the bath is initially polarised by applying $N = 50$ polarisation steps, and then, after a time period $t$, the polarisation of the bath is measured again [Fig. 4(a-iii)]. By performing the measurement while still at the NV-$^{13}$C resonance, we obtain a decay of $T_1^\text{pol} = 250\pm50\mu s$ [Fig. 4(d), orange data]. The decay time of this polarisation is limited by the NV spin $T_1^\text{NV} (50 \pm 5 \mu s$ on resonance). While the inner core is polarised and no longer interacting with the NV spin, the outer shell which is unpolarised can interact with the NV during the probe time $t$, causing a drastically shortened NV $T_1$. Once the NV has decayed, it acts as an additional noise source interacting with the polarisation of the spin bath [Fig. 4(e), bottom left]. This effect can be suppressed by initialising the NV into the |−1⟩ state before the evolution time [Fig. 4(a-iv)], effectively turning off the NV-$^{13}$C interaction [Fig. 4(e), bottom right]. Including this NV spin flip extends the $T_1$ of the bath to the order of 10 ms [Fig. 4(d), blue], which is limited by the phonon limited $T_1$ of the NV, on the order of milliseconds.\textsuperscript{29} These results agree well with theory [Fig. 4(d), solid lines], see supplementary material for details. We note that the acquisition

![FIG. 3. (a) Typical recording of the room temperature vs time, measured with the thermistor attached to the magnet, in the absence of active temperature control. (b) Variations in the NV frequency near the GSLAC, without temperature control (orange) showing a strong anti-correlation with (a), with first order stabilisation (green), and with second order stabilisation (blue). In the measurements leading to the green and blue curves, the room temperature fluctuations were similar to (a), see supplementary material.](103103-3_Broadway et al. Appl. Phys. Lett. 112, 103103 (2018))
time for each curve in Fig. 4(d) was ~20 h, during which the resonance condition was maintained; this would not have been possible to achieve without active temperature control with our apparatus.

We now compare our temperature-controlled permanent magnet to alternative approaches. Reaching the required magnetic field strength (e.g., ~0.1 T in this work) is very challenging using a standard electromagnet or solenoid, due to the restricted access to the diamond sample in typical confocal microscopes which employ bulky high numerical aperture objective lenses in quasi-contact with the sample and to heating induced by the large currents required. Ignoring heating-related drifts and instabilities, the stability of the magnetic field generated by an electromagnet is ultimately limited by the stability of the current source. Commercially available current sources generally feature a long term relative stability of the order of $10^{-3} - 10^{-4}$ for currents in the Ampere range and a setting resolution and noise level of the same order. For an NV centre near the GSLAC, this translates into 300 kHz to 3 MHz fluctuations (long term and noise) and setting resolution. In contrast, in this work, we obtained fluctuations as low as 100 kHz (200 kHz peak to peak), a 26 kHz setting resolution, and a noise negligible compared to the environmental noise.\footnote{A. Morello, J. J. Pla, F. A. Zwanenburg, K. W. Chan, K. Y. Tan, H. Huebl, M. Möttönen, C. D. Nagroho, C. Yang, J. A. van Donkelaar, A. D. C. Alves, D. N. Jamieson, C. C. Escott, L. C. L. Hollenberg, R. G. Clark, and A. S. Dzurak, Nature 467, 687 (2010).}

Thus, our approach already outperforms the electromagnet-based solution, for a fraction of the cost. Moreover, it could be further improved—by more than an order of magnitude—by using a permanent magnet material with a lower $\zeta_0$ (see supplementary material).

In summary, a temperature control scheme was implemented on a permanent magnet and was shown to be capable of performing magnetic field sweeps to produce CR spectra of electron and nuclear spins. Controlling the magnetic temperature improved the long term stability of the NV spin frequency from $\pm1$ MHz to $\pm100$ kHz and could be further improved on with different magnet materials. With the increased stability, dynamics of the $^{13}$C polarised bath were explored. These results indicated that for small polarised regions, the NV spin can significantly limit the lifetime of the polarisation imparted onto the bath. However, this effect can be mitigated by initialising the NV into a non-interacting state. This work shows that the simple addition of temperature control to a permanent magnet can overcome some of its shortcomings and outperform electromagnets in terms of precision and stability. Beyond laboratory experiments, this method can be extremely useful to realise portable devices based on NV centres or other spin qubit systems.

See supplementary material for more details on the magnet stabilisation procedure and the theory of the polarised bath evolution.


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