Quantum noise spectroscopy of non-monotonous spectra

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Understanding the physical origin of noise affecting quantum systems is important for nearly every quantum application. While quantum noise spectroscopy has been employed in various quantum systems, such as superconducting qubits and trapped ions, traditional spectroscopy methods are usually efficient in measuring noise spectra with mostly monotonically decaying contributions. However, there are important scenarios in which the noise spectrum is broadband and non-monotonous, thus posing a challenge to existing noise spectroscopy schemes. Here, we compared several methods for noise spectroscopy: spectral decomposition based on the CPMG sequence, the recently presented DYSCO sequence and a modified DYSCO sequence with a Gaussian envelope (gDYSCO). The performance of the sequences is quantified by analytic and numeric determination of the frequency resolution, bandwidth and sensitivity, revealing a supremacy of gDYSCO to reconstruct non-trivial features. Utilizing an ensemble of nitrogen-vacancy centres in diamond coupled to a high density \textsuperscript{13}C nuclear spin environment, we experimentally confirm our findings. The combination of the presented schemes offers potential to record high quality noise spectra as a prerequisite to generate quantum systems unlimited by their spin-bath environment.

Quantum systems are inherently subject to noise originating from their coupling to the environment, which in turn affects their coherence properties, with implications to quantum information processing, many-body dynamics and quantum sensing \cite{1,6}. As a consequence, studying the noise sources affecting quantum systems and optimizing schemes for mitigating them has been of great interest over the past decade \cite{2,9,5,11}. Moreover, devising new quantum noise spectroscopy methods (general techniques for analysing noise sources using quantum probes) is a fundamental aspect of the field and has attracted significant attention over the past years\cite{7,10–13}.

Quantum noise spectroscopy has advanced in recent years, and has been employed in the context of various quantum systems, such as superconducting qubits \cite{7}, trapped ions \cite{13}, \textsuperscript{13}C atoms in adamantane \cite{15}, optically trapped ultra-cold atoms \cite{16} and nitrogen-vacancy (NV) centres in diamond \cite{10,12}. These studies have led to a deeper understanding of the physical origins and dynamics of the noise sources and of the system-environment interaction (e.g. \cite{7,10,12}), as well as to advanced sensing applications (e.g. \cite{13}).

In most of the previous works, the spectrum of the relevant environmental noise was either nearly monotonically decreasing (e.g. $1/f$ noise \cite{7}, DC-centred Lorentzian \cite{12}), bandwidth-limiting techniques were employed \cite{12} or the noise spectroscopy was performed using straightforward continuous driving, resulting, again, in limited bandwidth (e.g. \cite{14}). These approaches are therefore limited in addressing important situations commonly encountered in realistic systems, in which the contributing noise is distributed over a large frequency bandwidth but the noise spectrum also exhibits strong resonant features (and thus a non-monotonous spectrum).

Here, we investigate the potential of noise spectroscopy based on three microwave (MW) driving sequences: the CPMG sequence post-processed by spectral decomposition (CPMG SD) \cite{10,12}, and the two recently introduced sequences: DYnamic Sensitivity COntrol (DYSCO) and a modified DYSCO scheme with a Gaussian envelope (gDYSCO) \cite{17}. The properties of the sequences are studied analytically and numerically in terms of accessible bandwidth, frequency resolution and gain as well as their implications for the reconstruction of noise spectra. Experiments were conducted utilizing an ensemble of NV centres in diamond coupled to a bath of \textsuperscript{13}C nuclear spins to confirm the predicted behavior.

In the last decade, the NV colour centre in diamond has emerged as a leading platform for quantum information, quantum meteorology and magnetic sensing \cite{1,18–24} due to its remarkable properties such as long coherence times at room temperature \cite{25,26}. Methods adopted from the field of nuclear magnetic resonance, such as dynamical decoupling \cite{27,30}, increased the coherence time even further \cite{26}. Substantial research efforts are invested in order to better understand the relevant noise sources affecting NV centres and their physical origins, as well as to optimize protocols to suppress their adverse effects \cite{10,15,31}.

The NV centre is a defect in the diamond lattice, in which one of the carbon atoms is replaced with a nitrogen atom and an adjacent site is substituted by a vacancy. A zero-field splitting of $\sim 2.87$ GHz between $m_s = 0$ and $m_s = \pm 1$ defines the triplet ground state. By applying a static magnetic field one can break the degeneracy between the $m_s = \pm 1$ sub-states through the Zeeman effect, thus creating an effective two-level system. The
NV centre electronic spin can be initialized and detected optically due to state-dependent fluorescence, and coherently manipulated within the ground-state spin manifold using MW fields \cite{20}.

The system’s decoherence due to environment-induced noise can be expressed as $C(t) = e^{-\chi(t)}$, where $\chi(t)$ contains the dependence of the decoherence processes on the spectral noise density $S(\omega)$ through \cite{32}:

$$
\chi(t) = \int_0^\infty \frac{d\omega}{\pi} S(\omega) \frac{F(\omega t)}{\omega^2} \tag{1}
$$

Here $F(\omega t)$ is the sequence dependent frequency filter function which is defined as the absolute square of the Fourier transform of the time-dependent sequence sensitivity function. For convenience, we define the filter function (FF) as: $\text{FF}(\omega t) = \frac{2F(\omega t)}{\pi t^2}$ such that Eq. (1) becomes $\chi(t) = \frac{1}{2} \int_0^\infty d\omega S(\omega) \text{FF}(\omega t)$.

Generally, the system’s sensitivity can be modulated in a pulsed or continuous manner. While pulsed sequences (e.g. CPMG) flip the sensitivity between ±1, the continuously driven DYSCO scheme enables arbitrary modulation of the sensitivity. Precise driving of the quantum sensor enables tuning of the FF in order to measure the noise environment at well defined frequencies $f_0$. The full noise spectrum $S(\omega)$ can be reconstructed out of a set of measurements \cite{32}.

One of the simplest pulsed sensitivity modulation sequences is the Hahn-Echo sequence \cite{33}, in which a single $\pi$ pulse is applied in the middle of the sequence of length $t_{\text{Hahn}} = 2\tau_{\text{free}}$ in order to refocus the NV spin and eliminate DC effects. Additionally, any noise contribution whose correlation time is shorter than the free evolution time $\tau_{\text{free}}$ cancels out. As an extension of the Hahn-Echo experiment, the CPMG-N pulse sequence \cite{24} consists out of $N$ equally spaced $\pi$ pulses with $2\tau_{\text{free}}$ time intervals between them (Fig. 1). By changing the free precession time $\tau_{\text{free}}$, the experiment time $t_{\text{CPMG}}$ and the number of pulses $N$, the sensing frequency can be adjusted according to:

$$
f_0^{\text{CPMG}}(\tau_{\text{free}}) \approx \frac{N}{2t_{\text{CPMG}}} = \frac{1}{4\tau_{\text{free}}} \tag{2}
$$

The total experiment time is given by $t_{\text{CPMG}} = 2N\tau_{\text{free}}$ and is ultimately limited by $2T_1$ \cite{35}. However, the pulsed nature of the sequence, i.e. stepwise sensitivity modulation (Fig. 2(a)), introduces higher harmonics into the filter function (Fig. 2(b)). A significant contribution to the decoherence can originate from these higher harmonics if the noise spectrum is not monotonically decreasing. While the spectral decomposition scheme incorporates precise knowledge of the FF, the reconstructed spectrum nevertheless suffers from artifacts of this origin. (See supplementary materials for a complete description of the spectral decomposition method).

The DYSCO pulse sequence was first presented by Lazari\v{c} et al. \cite{17} as a means for selective radiofrequency (RF) spectroscopy using NV centres. Contrary to spin-flipping sequences, DYSCO allows for control of the instantaneous sensitivity of the spin-sensor by precise pulse phase handling. This is achieved at the cost of permanent driving and reduction of the maximal sensitivity by a factor of $\frac{1}{2}$. Modulating the sensitivity function in a sinusoidal manner (Fig. 2(a)) results in a filter function free of higher harmonics. However, given the finite experiment time, the DYSCO filter function is a squared sinc function which has small side lobes (Fig. 2(b)). This effect can be suppressed by adding a Gaussian envelope to the sensitivity modulation (gDYSCO) (Fig. 2(a)), which removes the side lobes at the cost of a further reduced sensitivity and a slightly wider main peak (Fig. 2(b)).

For a more quantitative analysis of the FFs, we examined four parameters: resolution, minimum frequency $f_{\text{min}}$, maximum frequency $f_{\text{max}}$ and the gain $\Sigma$ (which is a measure for the system’s response at the desired frequency). While $f_{\text{min}}$ and $f_{\text{max}}$ define the system’s bandwidth, we introduce the FWHM of the FF main peak as a quantitative measure of the frequency resolution. However, it should be noted that when the FF contains side lobes (CPMG and DYSCO) they limit the resolution further by increasing the effective envelope around the main peak, so the FWHM only gives a bound on the resolution.

The DYSCO FF is a squared sinc function such that the FWHM$_{\text{DYSCO}} \approx \frac{0.884}{t_{\text{DYSCO}}}$ (calculated numerically), where $t_{\text{DYSCO}}$ is the the experiment time, limited by $T_{1,\rho}$ (the relaxation time in the driven system). On the other hand, the gDYSCO FF is a Gaussian with FWHM$_{g\text{DYSCO}} = \sqrt{\ln 2} \approx 0.655$, where $\sigma$ is the width of the time-domain Gaussian envelope. $\sigma$ is limited by the experiment time, and in our experiments and simulation it was chosen to be $\sigma = \frac{\ln 2}{\tau_{\text{free}}}$ such that the envelope is $\to 0$ at the beginning and end of the sequence. This results in a full width half maximum of
recall that the time scale in DYSCO is limited by \( T_{\text{CPMG}} \) numerically. The width is given by \( \text{FWHM} \approx 1.59 \tau_{\text{CPMG}} \). In contrast, the CPMG FF main peak frequency and FWHM can only be derived numerically. It is important to note that there is a significant difference between \( t_{\text{CPMG}} \) and the DYSCO experiment time \( t_{\text{DYSCO}} \). While for DYSCO, \( t_{\text{DYSCO}} \) is, due to its continuously driven nature, limited by \( T_1^\rho \) and remains constant regardless of the sensing frequency, in CPMG, \( t_{\text{CPMG}} \) changes with the sensing frequency which is given in Eq. 2. It follows that the FWHM can also be written as: \( \text{FWHM}_{\text{CPMG}} \approx 1.77 \tau_{\text{CPMG}} \) or \( \text{FWHM}_{\text{CPMG}} \approx 0.89 \tau_{\text{free}} \). In addition, \( t_{\text{CPMG}} \) is limited by \( T_2 \) (the transverse, spin-spin, relaxation time), which depends on \( N \) and is ultimately limited by \( T_1 \) (the longitudinal, spin-lattice, relaxation time). These non-trivial relations complicate the comparison between DYSCO and CPMG, but generally it can be stated that CPMG can achieve a higher resolution (narrower FF) for a given frequency by increasing the number of pulses.

The gain \( \Sigma \) is defined by the integral over the main peak in the region of the FWHM:

\[
\Sigma = \int_{f_0-\Delta}^{f_0+\Delta} FF(f) df \tag{3}
\]

Where \( f_0 \) is the FF peak frequency and \( 2\Delta \) is the FWHM. \( \Sigma \) is a measure of how much the coherence curve is affected by the presence of noise around the sensed frequency. While \( \Sigma \) varies slightly as a function of frequency \( f_0 \), overall for DYSCO it is approximately 60% \cite{17} of the CPMG gain \( \Sigma_{\text{CPMG}} \), and for Gaussian DYSCO it is about 20%.

CPMG has a clear advantage in terms of bandwidth, or dynamic range. The maximal frequency for CPMG is limited by the requirement \( 2\tau_{\text{free}} \gg \tau_\pi \), where \( \tau_\pi \) is the duration of a \( \pi \)-pulse. As a consequence, the maximal frequency is given by \( f_{\text{CPMG}} \ll f_{\text{Rabi}} \) (with \( f_{\text{Rabi}} \) being the Rabi/driving frequency). On the other hand, in DYSCO, as well as in gDYSCO, the maximal frequency is limited by the quantization of the sensitivity sine function (due to discrete phase steps). Quantizing the sine to \( n \) steps, where \( n \gg 1 \), leads to \( f_{\text{max}} \approx \frac{1}{2n}\pi \). Even though the expressions for \( f_{\text{max}} \) are similar, it needs to be stated that for small \( n \) the DYSCO sensitivity modulation approaches the step like case of CPMG. The reduced gain, \( \Sigma \), in combination with the appearance of higher harmonics in small \( n \) DYSCO reveals supremacy of CPMG SD to reconstruct high frequency noise. The minimum frequency \( f_{\text{min}}^{\text{CPMG}} \) for CPMG is estimated in a similar manner and is \( f_{\text{min}}^{\text{CPMG}} \approx \frac{1}{2\tau_{\text{free}}} \) (see supp. material for derivation). For DYSCO and gDYSCO the minimum frequency is given by the demand to fit at least one full period of the sensed frequency in the total experiment time so \( f_{\text{min}}^{\text{DYSCO}}>1 \Rightarrow f_{\text{DYSCO}}^{\text{min}} \gg \frac{1}{t_{\text{DYSCO}}} \).

Recall that the time scale in DYSCO is limited by \( T_1^\rho \).

Numerical simulations demonstrate the limitations of the different methods. As a proof of principle, the trivial case of a monotonically decreasing noise spectrum \( S(\omega) \) is considered. By integrating Eq. 1 with a set of CPMG FFs (see supp. material), complete coherence curves are simulated. Adding normally distributed noise to the coherence curves generates realistic data sets (inset of Fig. 3a) which are subsequently analysed by the CPMG SD method in order to reconstruct the spectrum. The results plotted in Fig. 3a show that for a monotonically decreasing noise spectrum the CPMG SD method reconstructs the noise spectrum with high precision. Repeating the procedure for the DYSCO FF reveals the noise spectrum with similar accuracy. In the next step a Gaussian peak around 62 kHz is added to the initial noise spectrum, which simulates a \( ^{13}\text{C} \) Larmor peak originating from surrounding nuclear spins at an external magnetic field of \( |B| \approx 60 \text{ Gauss} \). In Fig. 3b-d) the reconstruction of the simulated noise spectra with CPMG, DYSCO and gDYSCO is shown. Red data are obtained from noise free simulated coherence curves whereas green data points incorporate the simulated noise. The Gaussian DYSCO scheme results in a superior reconstruction of the \( ^{13}\text{C} \) peak, while CPMG suffers from a significant systematic bias caused by the higher harmonics and side lobes of the FF. When statistical noise is added, the limited sensitivity of gDYSCO alters the results, but the peak is still clearly visible as most of the noise affects the monotonous “background”. In the CPMG case, the noise has a limited effect on this “background”, but significantly flattens the noise peak. Also, it can be seen that the CPMG SD method, in the presence of noise, pushes the peak to slightly higher frequencies. This is caused by the asymmetry of the filter function and the higher harmonics.
The simulated noise reconstructed with gDYSCO reproduces the noise peak with much higher precision than DYSCO. Despite the fact that the main peak of the DYSCO FF is a factor of two narrower than that of the gDYSCO, it still produces a much wider $^{13}$C Larmor peak which is comparable to the one reconstructed by the CPMG SD method. This indicates a significant contribution originating from the side lobes of the DYSCO FF and highlights the fact that the resolution is not limited by the simple FF FWHM in the case of non-monotonic spectra. Nevertheless, the FWHM gives a bound on the resolution which is reached when the spectrum does not have a significant peak. Since both the CPMG FF and the DYSCO FF have feature-comparable side lobes, the resulting spectrum around the $^{13}$C Larmor peak appears similar for both of them, with the CPMG FF higher harmonics contributing as a 2$^{nd}$ order effect.

In conclusion, we have analysed and compared different quantum noise spectroscopy methods for the case of non-monotonic spectra, specifically containing a large and narrow resonant feature. We found that the gDYSCO scheme has better resolution and allows for a more precise detection of distinct features, while spectral decomposition based on CPMG exhibits higher sensitivity and larger bandwidth. Therefore, this work suggests that the best approach is a combination of different approaches in order to reveal full spectral information of non-trivial noise baths (our results indicate that optimized conditions might increase the sensitivity of gDYSCO, as described in the supplementary material, but this will be studied in detail in future work). This insight could provide an important tool for the study and characterisation of a wide range of quantum systems, such as various solid-state defects, super-conducting circuits, quantum dots and trapped ions, leading to a deeper understanding of the relevant physical processes, as well as optimized control schemes for quantum applications such as sensing and quantum information processing. In particular, this

|               | CPMG  | gDYSCO |
|---------------|-------|--------|
| $f$ [kHz]     | 60.2 ± 4.2 | 64.3 ± 0.5 |
| Width [kHz]   | 29.1 ± 2.2 | 13.8 ± 0.8 |

TABLE I. $^{13}$C Larmor frequency as extracted from the reconstructed experimental noise spectra by means of a Gaussian fit. The width of the Larmor feature is the fitted Gaussian 1σ.
method could be used in NMR/MRI measurements, traditionally done with pulsed XY8 sequences, to improve the sensitivity, bandwidth and accuracy. Other works that have found non-monotonous magnetic noise, such as pulsed modulation measurements done with ions\cite{Kotler2005} could also benefit from these new methods.

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g\text{DYSCO} does not reveal the noise spectrum (Fig. 3b). In addition, it can be clearly seen that the limited sensitivity of g\text{DYSCO} does not reveal the noise spectrum when its power is below $\approx 10^7$ Hz.

![FIG. 4. (a) Experimental coherence curves for CPMG-N experiments. Short time scales are densely sampled in order to reveal the $^{13}$C collapse. The revival points are also sampled in order to extract the decoherence curve. The envelope functions $\exp(- (x/T)^p)$ fitted to the revival points are shown as solid lines. (b) Coherence curve obtained from a g\text{DYSCO} measurement as a function of the modulation frequency. (c) The reconstructed spectrum using CPMG 2-128 and the spectrum obtained from the Gaussian DYSCO measurements. Both spectra were fitted with a Gaussian, the fitted parameters can be seen in Table 1. The $^{13}$C Larmor peak reconstructed by g\text{DYSCO} is narrower than the one obtained by CPMG SD, as predicted from the simulation results (Fig. 3)].
[24] L. Rondin, J.-P. Tetienne, S. Rohart, A. Thiaville, T. Hingant, P. Spinicelli, J.-F. Roch, and V. Jacques, *Nature Communications* 4, 2279 (2013).

[25] M. W. Doherty, N. B. Manson, P. Delaney, F. Jelezko, J. Wrachtrup, and L. C. Hollenberg, *Physics Reports* 528, 1 (2013).

[26] N. Bar-Gill, L. M. Pham, A. Jarmola, D. Budker, and R. L. Walsworth, *Nature Communications* 4, 1743 (2013).

[27] G. de Lange, Z. H. Wang, D. Ristè, V. V. Dobrovitski, and R. Hanson, *Science* 330, 60 (2010).

[28] B. Naydenov, F. Dolde, L. T. Hall, C. Shin, H. Fedder, L. C. L. Hollenberg, F. Jelezko, and J. Wrachtrup, *Physical Review B* 83, 081201 (2011).

[29] J. H. Shim, I. Niemeyer, J. Zhang, and D. Suter, *Physical Review Letters* 99, 40004 (2012).

[30] C. A. Ryan, J. S. Hodges, and D. G. Cory, *Physical Review Letters* 105, 200402 (2010).

[31] M. Kim, H. Mamin, M. Sherwood, K. Ohno, D. Awschalom, and D. Rugar, *Physical Review Letters* 115 (2015), 10.1103/PhysRevLett.115.087602.

[32] L. Cywiński, R. M. Lutchyn, C. P. Nave, and S. Das Sarma, *Physical Review B* 77, 174509 (2008).

[33] E. L. Hahn, *Phys. Rev.* 80, 580 (1950).

[34] S. Meiboom and D. Gill, *Review of Scientific Instruments* 29, 688 (1958).

[35] R. de Sousa, in *Electron Spin Resonance and Related Phenomena in Low-Dimensional Structures*, Topics in Applied Physics, Vol. 115, edited by M. Fanciulli (Springer Berlin Heidelberg, 2009) pp. 183–220.