# Supplementary Information: Decoherence of $V_B^-$ spin defects in monoisotopic hexagonal boron nitride

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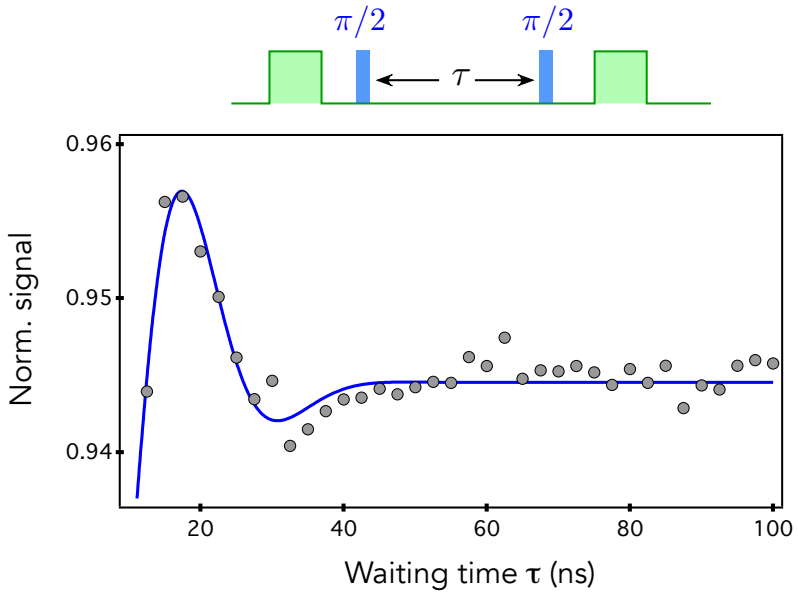
## Supplementary Note 1. Cluster correlation expansion calculations

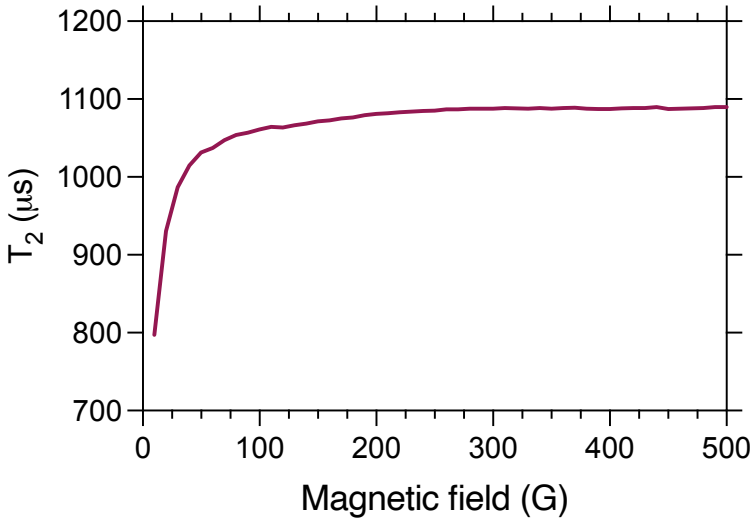
In order to theoretically study the isotope abundance dependence of the coherence time of the $V_B^-$ center in hBN, we implement the generalized cluster correlation expansion method (gCCE) [1,2], which is the state-of-the-art computational method to describe decoherence of point defect qubits coupled to a bath of nuclear spins. Before applying the method for calculating the coherence time of the $V_B^-$ center, we test our implementation on prevision results published in the literature for the Hahn echo coherence time of the NV center in diamond. As can be seen in Supplementary Figure 2, our simulation reproduces the magnetic field dependence of the coherence time reported in Ref. [3].

In our study, the overall coherence function ($C$) is obtained as a product of the zero ($C_0$), first ($C_1$), second ($C_2$), and third ($C_3$) order coherence contributions in gCCE-3 approximation [1,2]. In Supplementary Figure 3, we depict the time dependence of the coherence functions of different orders. As can be seen, the first and second order coherence functions dominate the decay, however, the third order contributions may not be neglected as well.

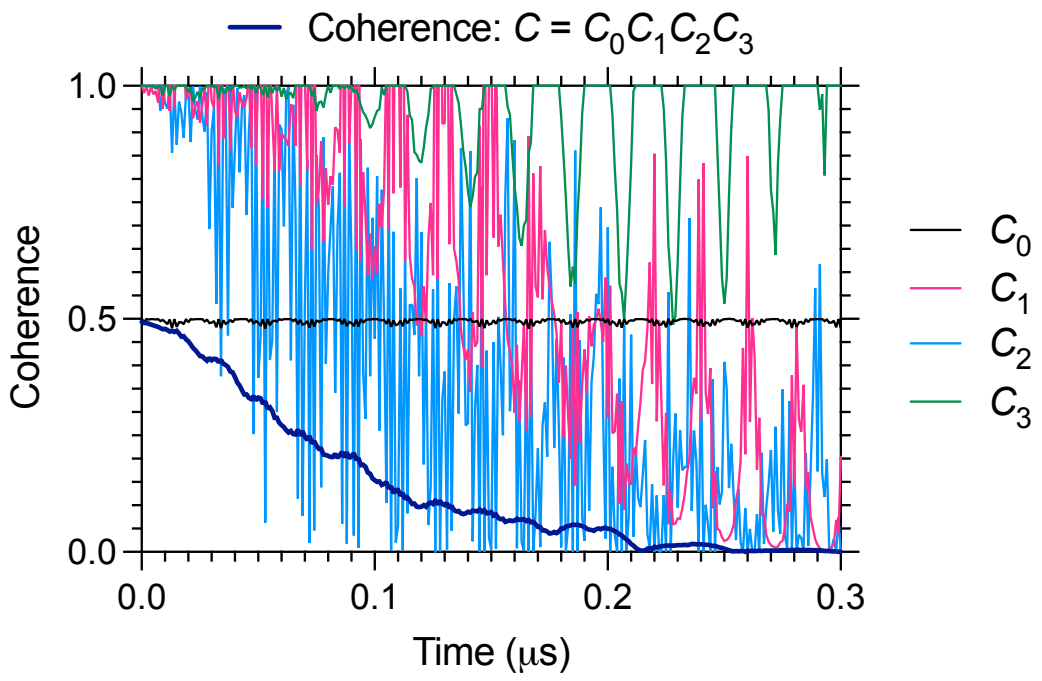
Nuclear spins and nuclear spin pairs at different distances from the $V_B^-$ center have different effects on the coherence function. Since the dipole-dipole interaction rapidly decreases with the distance of the spins, a proper cut-off radius can be found in general, beyond which the nuclear spins have negligible influence on the coherence properties of the $V_B^-$ center. In our implementation, we define three cut-off radii, see Supplementary Figure 4(a). The first order contribution $C_1$ accounts only for pair interactions. Convergence of the coherence time with respect to the maximal distance of the electron spin and the nuclear spins is depicted in Supplementary Figure 4(b). As can be seen at $r_1 = 10$ Å the first order contribution converges. The second order coherence contribution $C_2$ is obtained from three-spin clusters that can account for nuclear spin-nuclear spin flip-flops. Here we define two cut-off radii, $r_2$ and $d_2$, that define the maximal distance of the nuclear spins from the $V_B^-$ center and the maximal distance of the nuclear spins in the second order clusters. In Supplementary Figure 4(c,d) we show how the $T_2$ time converges with respect to these parameters. Convergent results are obtained for $r_2 = 7.5$ Å and $d_2 = 10$ Å. In addition, the first and second neighbor nuclear spins give rise to four body correlation effects in the third order coherence contribution $C_3$, however, they have only secondary contribution to the decoherence of the $V_B^-$ center in hBN [see Supplementary Figure 3].



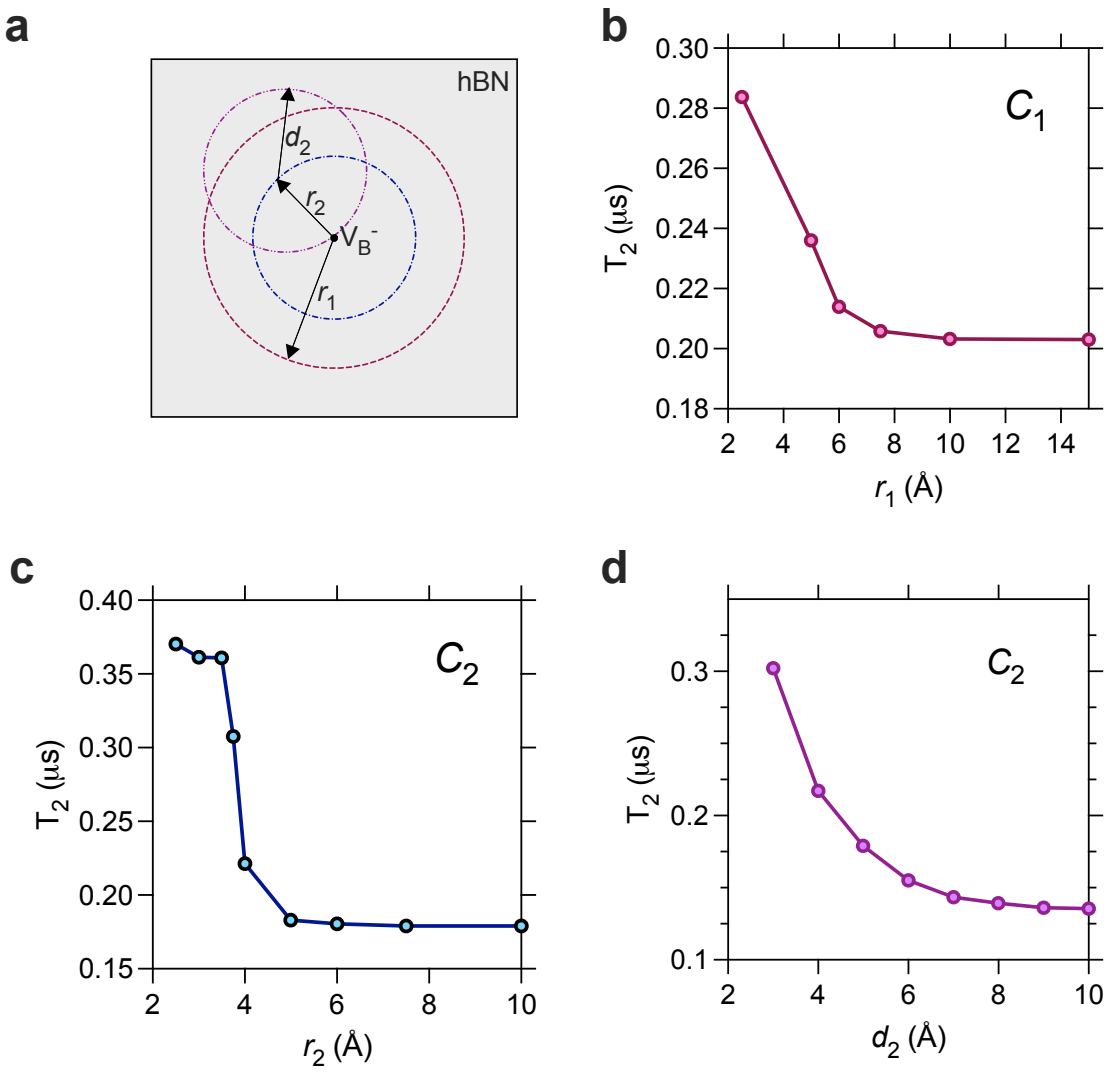

**Supplementary Figure 1.** Typical free induction decay of an ensemble of $V_B^-$ spin defects hosted in the h$^{10}$BN crystal, showing a spin dephasing time $T_2^\star \sim 20$ ns. The measurement is carried out under the same experimental conditions as those used to record the data shown in Figure 3 of the main article.




**Supplementary Figure 2.** Magnetic field dependence of the Hahn echo coherence time of the NV center in diamond.




**Supplementary Figure 3.** Different order contributions to the decay of the coherence function of the $V_B^-$ center in hBN.




**Supplementary Figure 4.** Convergence of the coherence time with respect to different cut-off radii of the CCE method. **a** Definition of the different cut off radii. Dependence on **b** the cut off radius $r_1$ of the first order CCE contribution $C_1$ and **c** the cut off radius $r_2$ of the second order CCE contribution $C_2$. **d** Convergence of the coherence time with respect to the maximal distance $d_2$ of the nuclear spins in the second order contribution.

## Supplementary References

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