Introduction.— Defects in solids possessing spin are promising solid-state qubits whose long spin coherence times permit optical initialization and readout [1, 2]. The energy levels of spin-1 centers lacking inversion symmetry, such as NV centers in diamond, shift in response to electric fields to first order, motivating their application to quantum sensing and metrology of charge dynamics [3–16], including for nanoscale probes of many-body physics [17–20]. Undesirable fluctuating electric and magnetic fields also couple to the spin [5, 12, 17–21, 31], and their effects increase near the surface due to trapped charges and imperfections. The differing spatial dependence of fluctuating electric fields produced by uncorrelated monopole (point-like) and dipole charges at the surface, \( E \propto |r|^{-2} \) and \( E \propto |r|^{-3} \), produce a surface noise spectral density \( S_E(\omega) \propto d^{-2} \) and \( S_E(\omega) \propto d^{-4} \) [12, 24, 25, 31], where \( d \) is the spin center depth; this noise at a resonant frequency for spin transitions leads directly to spin relaxation and decoherence and reduces both the sensitivity to external fields and the optical contrast in readout. The rapid increase in noise near a surface conflicts with the desire to place the spin as close to the surface as possible to increase sensitivity to a signal originating from outside the material and to improve the sensor’s spatial resolution. A major focus for quantum sensing, therefore, explores techniques to reduce these noise sources [21, 22, 24, 32, 33]; similar goals help advance defect-based hybrid quantum systems [34–36], as the coupling of a spin to another excitation (e.g., magnon, phonon) also improves the closer the spin comes to the surface. These efforts depend on an accurate understanding of the noise and its depth-dependent scaling.

Here we show that very different depth \((d)\) dependencies for the frequency \((\omega)\) dependent noise spectral density \( S_E(\omega) \) emerge when spatial and time correlations are considered; our general formalism relies on the two-point correlation functions of the fluctuating point-like charges’ surface density, \( \langle \delta \rho(r',t')\delta \rho(r,t) \rangle \), and the fluctuating electrostatic potential at the crystal surface, \( \langle \delta \phi(r',t')\delta \phi(r,t) \rangle \), yielding \( S_E^\rho \) and \( S_E^\phi \), respectively.

For example, for diffusive motion of surface charges we find \( S_E^\rho \propto d^{-4} \) instead of the \( d^{-2} \) dependence found for uncorrelated fluctuation of point-like particle densities. We find \( S_E^\rho \) is independent of depth when both (1) the fluctuating charge density spatial correlation falls off exponentially with distance (e.g., from crystal surface distortions and imperfections [47, 48]) and (2) the spin depth is much shorter than the correlation length.

Surface electric dynamics influences the quantum coherence of near-surface spin centers through spatial and temporal fluctuations of the surface charge density and the electrostatic potential at the crystal surface. The power law of the electric noise’s spectral density dependence on spin center depth is not solely determined by whether the surface charges fluctuate as monopoles or dipoles. The power instead depends sensitively on the structure of the surface charge and surface potential correlation functions. For surface dynamics originating from diffusion, the spin center’s relaxation and decoherence times exhibit a temporal crossover near the correlation time of the fluctuators and thus provide a quantitative fingerprint of the diffusive behavior of charged particles at surfaces.

![Electric noise](image-url)
We further obtain $S_{E}^{δ ρ} \propto d^{-4}$ for exponentially decaying spatial correlations of the surface potential fluctuations. In general, the depth-dependent behavior of $S_{E}^{δ ρ}$ and $S_{E}^{δ φ}$ strongly depends on the detailed physics of the surface noise phenomenon and on both $⟨δ ρ(r',t')δ ρ(r,t)⟩$ and $⟨δ φ(r',t')δ φ(r,t)⟩$. $S_{E}^{δ ρ}$ and $S_{E}^{δ φ}$ even exhibit different depth dependences for the same mathematical form for both $⟨δ ρ(r',t')δ ρ(r,t)⟩$ and $⟨δ φ(r',t')δ φ(r,t)⟩$. These results suggest a new sensing methodology: correlating surface-dependent physical phenomena with $d$ and $ω$ dependent noise spectra detected by electric-field sensitive quantum sensors; for spin centers sensing diffusive behavior the diffusion constants and correlation times can be extracted from detailed time-dependent measurements of spin coherence.

*Fundamentals of electric noise at the diamond surface.* — Hydrogen or oxygen termination shift the surface potential for diamond differently [49–52], similar effects emerge from imperfections of crystal termination [53] along with spatial fluctuations of the surface potential [54, 55]. Interaction of these levels with trapped charges in the nitrogen-doped diamonds that host NV centers result in the creation of an effective surface two-dimensional (2D) hole or electron gas [50, 53, 56–59]. These charges do not distribute uniformly and move around due to thermal fluctuations of the charge’s position, collision between charges, capture and ionization by donors and acceptors, and other processes. Moreover, as the read-out and initialization of the sensing spin requires laser illumination, the light increases these fluctuations. Thus the fluctuation of charged particles and the local surface potential produce noise that is experienced by shallow spins, causing decoherence and relaxation of the spin state.

The influence of these temporal fluctuations of charge density and surface potential on the spin center properties occurs via a temporally fluctuating electric field at the spin center position $r = r_d$, $E(r_d,t) = (E(r_d)) + δ E(r_d,t)$, which causes spin decoherence and relaxation [5, 12, 17, 21, 31] and increases the photoluminescence linewidth [52, 53, 60, 61]. For Dirichlet boundary conditions

$$E(r_d,t) = \frac{1}{\varepsilon} \int_V dr' K(r_d - r') ρ(r',t) + \oint_S dσ' φ(r_S',t) \frac{∂K(r_d - r_S')}{∂n'},$$  

and

$$⟨δ E_δ (r_d,t) δ E_μ (r_d,0)⟩ = \left( \frac{1}{\varepsilon} \right)^2 \int_V \int_V dr' dr'' K_μ(r_d - r') K_ν(r_d - r'') δ δ(ρ(r',t)δ ρ(r'',0)) + \oint_S dσ'' K_μ(r_d - r'') δ K_ν(r_d - r'') \frac{∂δ φ(r''_||,t)}{∂z} \frac{∂δ φ(r''_||,t)}{∂z} ⟨δ φ(r''_||,0)⟩.$$  

\section*{References}

We restrict fluctuations of $ρ$ and $μ$ to occur only on $S_δ$, thus $ρ(r,t) = q ϕ(z)n(r||,t)$, with $q$ the fluctuator’s charge. We also assume translational symmetry of the correlation function for the fluctuating surface density and surface potential along the surface, $Π_{δ ρ}(r''_|| - r||,t) = q^2 〈δ n(r''_||,t)δ n(r||,0)⟩$ and $Π_{δ φ}(r''_|| - r||,t) = 〈δ φ(r''_||,t)δ φ(r||,0)⟩$, respectively. Finally, $S_{E_δ}(ω) = \int_{−∞}^{∞} dt 〈δ E_δ (r,t) δ E_δ (r,0)⟩ e^−iωt$, yielding

$$S_{E_δ}^{δ ρ}(ω) = \left( \frac{1}{2\pi} \right)^2 \int \frac{dk}{(2π)^2} \mathcal{F}_{2D}^{k||}[K_ν(r_d)] \mathcal{F}_{2D}^{−k||}[K_ν(r_d)] \mathcal{F}_{2D}^{ω}[Π_{δ ρ}(r''_|| - r||,t)],$$  

$$S_{E_δ}^{δ φ}(ω) = \int \frac{dk}{(2π)^2} \mathcal{F}_{2D}^{−k||}[∂z K_ν(r_d)] \mathcal{F}_{2D}^{ω}[Π_{δ φ}(r''_|| - r||,t)].$$  

\section*{Acknowledgments}

For the 2D Fourier transform

$$\mathcal{F}_{2D}^{k||}[K_ν(r_d)] = \int dr || K(r_d)e^{−ik||r||},$$  

and

$$\mathcal{F}_{2D}^{k||}[K_ν(r_d)] = \int dr || K(r_d)e^{−ik||r||},$$  

we use the following notation:

\begin{align*}
\mathcal{F}_{2D}^{k||}[K_ν(r_d)] & = \frac{1}{2} \left[ i (δ_{ν,z} + δ_{ν,y}) k_ν \right] |k_|||e^{−|k_|||z}, \\
\mathcal{F}_{2D}^{k||}[∂z K_ν(r_d)] & = \frac{1}{2} \left[ i (δ_{ν,z} + δ_{ν,y}) k_ν \right] |k_|||e^{−|k_|||z},
\end{align*}
Equations (3)–(5) establish the relation between the electric spectral noise and the temporal and spatial Fourier transform of both $\Pi_{\delta p}(r'_\parallel - r_\parallel, t)$ and $\Pi_{\delta p}(r'_\parallel - r_\parallel, t)$.

Fluctuating surface density from diffusion — We assume $n(r_\parallel, t)$ satisfies

$$ \left( \frac{\partial}{\partial t} + \frac{1}{\tau} + \mu E_\parallel \cdot \nabla_\parallel - D \nabla_\parallel^2 \right) n(r_\parallel, t) = 0, $$

which includes diffusion, drift and a finite carrier lifetime. Accordingly, the corresponding Green’s function satisfies

$$ \left( \frac{\partial}{\partial t} + \frac{1}{\tau} + \mu E_\parallel \cdot \nabla_\parallel - D \nabla_\parallel^2 \right) G(r_\parallel, t) = \delta(r_\parallel) \delta(t) $$

and

$$ \delta n(r_\parallel, t) = \int d^3r' G(r_\parallel - r'_\parallel, t) \delta n(r'_\parallel, 0), $$

so $\Pi(r'_\parallel - r_\parallel, t) = \int d^3r' G(r'_\parallel - r_\parallel, t) \Pi(r'_\parallel - r_\parallel, 0)$. With this, Eq. (4) connects the noise’s spectral density to both the electrostatic Green’s function Kernel $K(r' - r)$ and the Green’s function describing the dynamics of our charged particles, $G$. For purely diffusive time evolution of the fluctuation correlator, $\langle \delta n(r'_\parallel, 0) \delta n(r_\parallel, 0) \rangle = \delta(r'_\parallel - r_\parallel) \langle n(r_\parallel, 0) \rangle$ and $\mathcal{F}_{2D,\omega}[\Pi(r'_\parallel - r_\parallel, t)] = \mathcal{F}_{2D,\omega}[G(r_\parallel, t)] n_S$ with $n_S = \langle n(r_\parallel, 0) \rangle$ for uniform density, and $\mathcal{F}_{2D,\omega}[G(r_\parallel, t)] = e^{i\omega t + i\mu E_\parallel \cdot k_\parallel + Dk_\parallel^2 t^{-1}}$ following from Eq. (7).

We now use this equation within Eq. (1) assuming first $E_\parallel = 0$ and $1/\tau \to 0$, i.e., purely diffusive motion, which for $\mu = x, y$ yields

$$ S_{E_\parallel}(\omega) = \left( \frac{q}{4\pi \varepsilon} \right)^2 n_S \pi \times \frac{2D}{\omega^2} \frac{d^2}{d\omega^2} \int_0^\infty \frac{e^{3e^{-2e}}}{1 + (\varepsilon^2 e^2)^2}, $$

where $\epsilon$ is a dimensionless variable and $S_{E_\parallel} = S_{E_x} \equiv S_E$. We cannot decouple the temporal part of the spectral noise from the spatial part. However, there are two limits, dictated by the value of $d^2\omega/D$, in which Eq. (8) can be solved analytically, yielding

$$ S_{E}(\omega) \approx \begin{cases} B_0 (3D/4d^4)^2 & d^2\omega/D \gg 1 \\ B_0 \left[ \ln \left( D/4d^4 \right) + 2\Gamma \right] / D & d^2\omega/D \ll 1 \end{cases} $$

with $B_0 = (q/4\pi \varepsilon)^2 n_S \pi$ and the Euler-Gamma number $\Gamma = 0.577$. In Fig. 1(a) we plot $S_{E}^{p}[\omega]$ as a function of $d/d_c$ with $d_c = (D/\omega)^{1/2}$. Although $S_{E}^{p} \propto d^{-4}$ for $d \gg d_c$, we obtain a $-\ln d$ dependence for $d \ll d_c$, with the intermediate gray region $0.1 \lesssim d/d_c \lesssim 10$ yielding $d^{-\gamma}$ (with $\gamma = 1, 2$ and 3) shown in Fig. 2(a).

A $d^{-4}$ depth dependence of the spin coherence time reported in the literature was attributed to the dipole character of the fluctuators [29, 31, 64]. Our results show that instead, this depth dependence can also be obtained from point-like charges that diffuse. In Fig. 2(b) we plot $S_{E}^{p}[\omega]$ as a function of $d$ for different diffusion coefficients. We note that for $d \ll 1$ nm faster diffusion (larger $D$) suppresses noise, similar to motional narrowing [55, 60]. However, for $d \gg 1$ nm we obtain the opposite behavior, and a plot of $S_{E}^{p}$ as a function of $D$ shows a maximum at $D \approx 1$ nm$^{-2}$ GHz for $\omega = 1$ GHz and $d = 1$ nm [Inset Fig. 2(b)]. We plot in Fig. 2(c) $S_{E}^{p}$ as a function of $\omega$ where we see the crossover between $\omega^2$ and $-\ln \omega$ at $\omega_c = D/d^2$.

Fluctuating surface density with exponential spatial correlation— An exponential correlation function of the fluctuating surface density, i.e., $\Pi(r'_\parallel - r_\parallel, t) = \chi(t) (n_S/A)e^{-|r'_\parallel - r_\parallel|/\lambda'}$ with $\ell > 0 \in \mathbb{Z}$ and $A$ the surface area, is common in crystals with inhomogeneous surfaces, and it can be obtained theoretically assuming random crystal surface distortions and imperfections [47, 48]. This behavior has also been experimentally observed with X-ray scattering from crystal surfaces [47, 48, 67, 68], which directly measures the degree of spatial correlation between fluctuations of the electronic density. Accordingly, $\mathcal{F}_{2D,\omega}[\Pi(r'_\parallel - r_\parallel, t)] = \chi(t) (n_S/A) \lambda^2 \exp(-\lambda^2 k_\parallel^2/4)$ for $\ell = 1$ and $\ell = 2$, respectively. Then from Eq. (5),

$$ S_{E}^{p,\ell=1}(\omega) = \left( \frac{q}{\varepsilon} \right)^2 \frac{n_S}{8A} \frac{F_{2D}[\chi(t)]}{(d/\lambda)^2} \int_0^\infty \frac{e^{-2e}}{1 + (\lambda/d)^2 e^2} d\epsilon, $$

$$ S_{E}^{p,\ell=2}(\omega) = \left( \frac{q}{\varepsilon} \right)^2 \frac{n_S}{8A} \frac{F_{2D}[\chi(t)]}{(d/\lambda)^2} \int_0^\infty \frac{e^{-2e}}{4(d/\lambda)^2} d\epsilon. $$

(10)
Only be accurately interpreted as due to the fluctuation

\[ S_E(\omega, d) \approx \begin{cases} 
B_1 F_{2D} \chi(t) & d \ll d_c, \\
B_1 F_{2D} \chi(t) / [4 (d/d_c)^2] & d \gg d_c,
\end{cases} \]

(12)

\[ S_E(\omega, d) \approx \begin{cases} 
B_1 F_{2D} \chi(t) & d \ll d_c, \\
B_1 F_{2D} \chi(t) / [8 (d/d_c)^2] & d \gg d_c,
\end{cases} \]

(13)

with \( B_1 = (q/e)^2 n_s/(8A) \). We see that both exponential correlation functions produce similar asymptotic behaviors, with a \( d^{-2} \) depth dependence for \( d \gg d_c = \lambda \). This depth dependence can also be obtained by taking the limit \( \lambda \to 0 \) leading to

\[ \langle \delta n(r_i', t) \delta n(r_j, 0) \rangle \propto f(t) \chi(t), \]

This represents a situation where uncorrelated fluctuations add incoherently. Thus a reported \( S_E \propto d^{-2} \) [12, 24, 25, 31] can only be accurately interpreted as due to the fluctuation of point-like charges for correlation lengths much smaller than the defect depth. As \( d \approx 10 \text{ nm} \), this implies that \( \lambda \lesssim 1 \text{ nm} \). Compared to Eq. (9), Eqs. (12) and (13) show that different classes of surface density spatial correlation function yield different depth dependences. In Fig. 2(d) we plot Eq. (10) as a function of \( d \) for \( \lambda = 1 \) and 10 nm, together with its asymptotic functions given by Eq. (12). Surprisingly, there is a plateau of \( S_E^{d_p} \) for depth much shorter than \( d_c = \lambda \), with nominal value independent of the power of the exponential correlation [See Fig. 1(b)]. This independence of depth can be understood through \( \lambda \to \infty \), which produces

\[ \langle \delta n(r_i', t) \delta n(r_j, 0) \rangle \propto f(t), \]

i.e., the fluctuations are strongly correlated and add coherently. We also see that for intermediate depth values, \( d \approx d_c \), \( S_E^{d_p} \) scales with \( d^{-1} \).

**Effect of the fluctuating surface potential.** — The fluctuation of the electrostatic potential cannot be described by the diffusive model Eq. (6). Alternative, we assume that the surface is composed of plaquettes [55, 69] with varying local potential, \( \phi_i(t) \) and \( \langle \phi_i(t) \rangle = \phi_0 \). Moreover, we assume \( \delta \phi \left( r_i', t \right) = \sum_{i=1}^{N} \delta \phi_i(t) \Theta \left( r_i \right) \) where
\( \Theta_i(\mathbf{r}_i) \) is the square function associated with the plaquette \( i \), i.e., it assumes value 1 (0) inside (outside) plaquette \( i \), centered at \( \mathbf{r}_i \) \( ^{[55, 69]} \), and with \( \int d\mathbf{r}_i \sum_i \Theta_i(\mathbf{r}_i) = A \).

Using \( \langle \delta \phi_i(t) \delta \phi_j(0) \rangle = \delta_{ij} \langle f_\phi(t) \rangle \), 
\( \sum_i \langle \Theta_i(\mathbf{r}_i) \Theta_i(\mathbf{r}_i') \rangle = N \langle \Theta_i(\mathbf{r}_\|) \Theta_i(\mathbf{r}_\|') \rangle \) due to the randomness of \( \Theta \), where \( N \) is the total number of plaquettes. Assuming spatial transitional symmetry of the correlation function of the plaquette positions, 
\( \langle \Theta_i(\mathbf{r}_\|) \Theta_i(\mathbf{r}_\|') \rangle = e^{-|\mathbf{r}_\| - \mathbf{r}_\|'|/A^\ell} \)
\( ^{[70]} \).

For \( \ell = 1 \) and \( \ell = 2 \), we obtain
\[
S_{E_x}^{\delta \phi, \ell=1}(\omega) = \frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle N A^2}{4d^4} \int_0^\infty \frac{dt}{1 + (\Lambda/d)^2} \frac{e^3}{(1 + (\Lambda/d)^2)^{3/2}}, \tag{14}
\]
\[
S_{E_x}^{\delta \phi, \ell=2}(\omega) = \frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle N A^2}{4d^4} \int_0^\infty \frac{dt}{1 + (\Lambda/d)^2} e^{-2\sigma/\Lambda^2}, \tag{15}
\]

where Eq. \( (14) \) appears in Ref. \( ^{[69]} \). The above integrals exhibit asymptotic analytical behavior given by
\[
S_{E_x}^{\delta \phi, \ell=1}(\omega) = \left\{ \begin{array}{ll} \frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle (N/16d\Lambda)}{d \ll \Lambda}, & \\
\frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle (3N A^2/6d^4)}{d \gg \Lambda} \end{array} \right. \]
\[
S_{E_x}^{\delta \phi, \ell=2}(\omega) = \left\{ \begin{array}{ll} \frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle (N A^2)}{d \ll \Lambda}, & \\
\frac{F_{2D}^{\omega} \langle f_\phi(t) \rangle (3N A^2/6d^4)}{d \gg \Lambda} \end{array} \right. \tag{16}
\]
producing a crossover between \( d^{-1} \) (\( d^0 \)) and \( d^{-4} \) depth dependences for \( \ell = 1 \) (\( \ell = 2 \)) as can be seen in Fig. \( 2(e) \) [Fig. \( 2(f) \)]. This result demonstrates that the dependence \( d^{-4} \) cannot be attributed solely to the electric noise arising from fluctuating dipole charges, and that the \( d \) dependence for short characteristic lengths \( \Lambda \) depends strongly on the spatial form of the correlation, i.e., \( e^{-|\mathbf{r}|/\Lambda} \) or \( e^{-|\mathbf{r}|^2/\Lambda^2} \). Figures \( 2(c) \) and \( 2(f) \) show \( d^{-1} \), \( d^{-2} \) and \( d^{-3} \) dependences for intermediate values of \( d \).

Finally, for both \( \ell = 1 \) and \( \ell = 2 \) we see that the bigger (smaller) the \( \Lambda \) the stronger (weaker) the suppression of the noise for \( d \ll \Lambda \) (\( d \gg \Lambda \)).

**Sensing the diffusion with spin defects.**—Our theory suggests sensing the diffusive regime using spin centers, and extracting the corresponding diffusion constant \( \mathcal{D} \). This could occur via multiple spin centers located at different depths. The crossover from \( d^{-4} \) to \( d^{-3} \) to \( d^{-2} \) to \( d^{-1} \) to \( -\ln d \) should be seen through both \( T_{1,1} \) and \( T_{1,2} \), with experimental results showing a trend similar to Figs. \( 1 \) and \( 2 \) as \( T_{1,1,2} \propto S_E(\omega) \). From the critical depth of the crossover, \( d_c \), the diffusion coefficient would be found through \( \mathcal{D} = d_c^2 \omega_c \). The diffusive regime could also be identified with only one spin center located at \( d \), by measuring \( 1/T_1 \) or \( 1/T_2 \) as a function of \( \omega_c \), which should follow a trend similar to Fig. \( 2(c) \) with a crossover located at \( \omega_c \), and a diffusion coefficient of \( \mathcal{D} = d^2 \omega_c \).

In analogy with recent work using spin centers to sense many-body interactions \( ^{[19, 20]} \), here we define the average probe coherence, \( \mathcal{C}(t) \), which characterizes the time evolution of both the dephasing or relaxation processes due to electric noise. For Gaussian noise \( \mathcal{C}(t) = e^{-\delta_t^2/2} \int_0^\infty dt' \int_0^\infty dt'' \langle E_{\omega}(t')E_{\omega}(t'') \rangle e^{i\theta(t'-t'')} \)
\( ^{[31, 33]} \) where \( \delta_t \) is proportional to the spin-defect electric dipole moment. \( \Omega \) characterizes either spin relaxation, with its value given by the corresponding frequency difference of the levels, or spin decoherence for \( \Omega = 0 \) \( ^{[31, 33, 34]} \). In terms of the electric noise spectral density
\[
\mathcal{C}(t) = \exp \left\{ -\frac{\delta_t^2}{2} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} S_E(\omega) \sin^2 \left( \frac{\omega + \Omega}{2\pi} t \right)^2 \right\}, \tag{18}
\]
with a limit \( \mathcal{C}(t) \approx \exp \left\{ -\frac{\delta_t^2}{2} S_E(t=0)t^2 \right\} \) with \( S_E(t=0) = \int_{-\infty}^{\infty} d\omega/(2\pi)S_E(\omega) \) for \( t \ll \tau_c \), and \( \mathcal{C}(t) \approx \exp \left\{ -\frac{\delta_t^2}{2} S_E(\omega = -\Omega)t \right\} \) for \( t \gg \tau_c \) with \( \tau_c = d^2/\mathcal{D} \). For diffusion the asymptotic behaviors are
\[
\mathcal{C}(t) = \left\{ \begin{array}{ll} \exp \left\{ -\frac{\delta_t^2}{2} S_E^\phi(0)t^2 \right\}, & t \ll \tau_c \\
\exp \left\{ -\frac{\delta_t^2}{2} S_E^\phi(-\Omega)t \right\}, & t \gg \tau_c \end{array} \right. \tag{19}
\]
which indicates a crossover between two different exponential decays, namely, \( e^{-\mathcal{D}t} \) and \( e^{-Bt} \). As \( \lim_{\omega \to 0} S_E^\phi(\omega) \to \infty \), this asymptotic behavior only holds for \( \Omega \neq 0 \). For \( \Omega = 0 \) we still find asymptotic behavior, but it is obtained by first performing the integral of Eq. \( (18) \) and then taking the limit \( \Omega \to 0 \). Thus decoherence and relaxation dominated by carrier diffusion can be demonstrated and observed experimentally through the time evolution of \( \mathcal{C}(t) \), shown in Fig. \( 3 \). This makes it possible to determine the diffusion correlation time as
\[ \tau_c = \tau_c(D). \] The depth dependence \( \propto d^{-2} \) is only obtained if \( t \ll \tau_c \), which causes the temporal decay of \( C(t) \) to be independent of \( D \); for short times diffusion does not influence the decoherence or relaxation of the spin center. For \( t \gg \tau_c \) the depth dependence is dictated by Eq. \[8\].

**Concluding remarks.**—We have developed a theory which predicts that the depth dependence of spin center decoherence and decay processes are strongly influenced by both the two-point correlation of the fluctuating surface particles’ density and the surface electrostatic potential. Our framework predicts that diffusive phenomena yield a non-trivial temporal behavior for the average probe coherence of spin centers, with a crossover between different exponential decay forms determined by the effective correlation time at the spin center. As a consequence, both the crossover point and the exponents contain a direct signature of diffusion, and permit extraction of the diffusion coefficient.

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