Kilohertz electron paramagnetic resonance spectroscopy of single nitrogen centers at zero magnetic field

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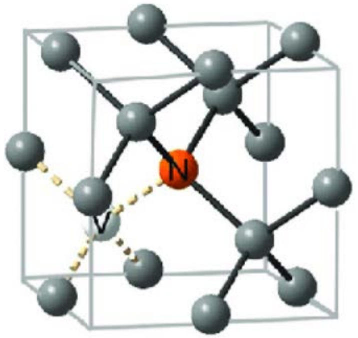
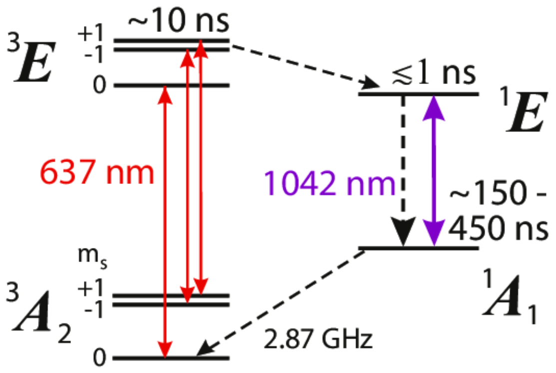
NV center–based detection

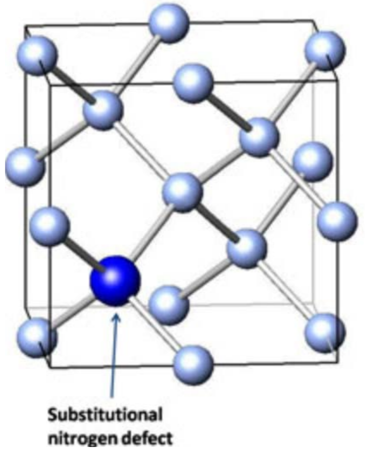
NV center
(**sensor**)

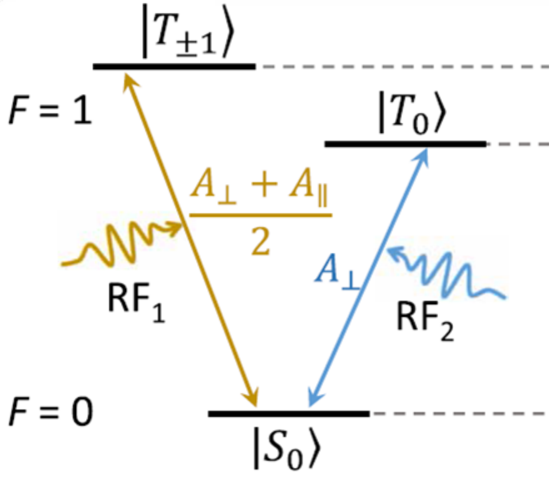




P1 center
(**target**)

$S = 1/2$, $I = 1/2$ system





$$H_t = \mathbf{S} \cdot \mathbf{A} \cdot \mathbf{I}$$

$$|T_{+1}\rangle = |\uparrow\uparrow\rangle,$$

$$|S_0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle),$$

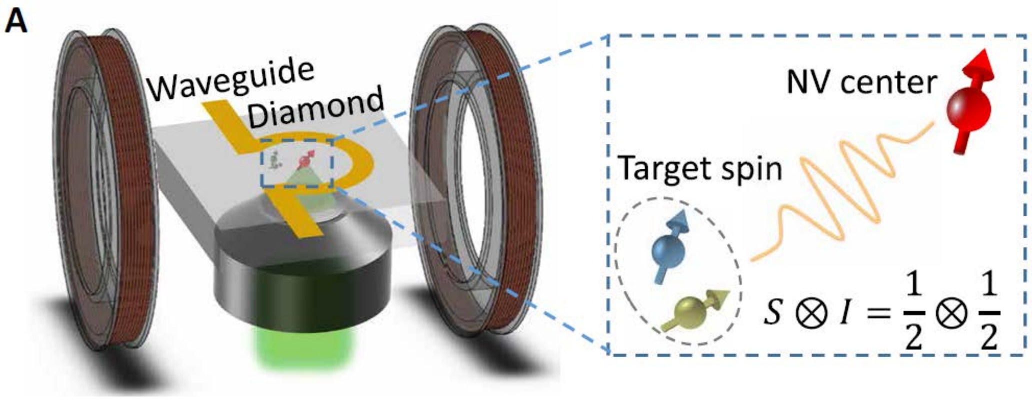
$$|T_0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle),$$

$$|T_{-1}\rangle = |\downarrow\downarrow\rangle,$$

$$H_{\mathrm{dd}} \approx \frac{\mu_0 \gamma_{\mathrm{NV}} \gamma_{\mathrm{tar}} \hbar}{4\pi r^3}(\cos\theta_{\mathrm{e}} - 3\cos\theta_r \cos\theta_{r'}) S_z^{\mathrm{NV}} S_{zz}^{\mathcal{T}}$$
$$= C(\theta_{\mathrm{e}}, r, \theta_r, \theta_{r'}) S_z^{\mathrm{NV}} S_{zz}^{\mathcal{T}},$$

NV center–based zero-field EPR spectrometer

$$H_0 = A_\perp(S_x I_x + S_y I_y) + A_\parallel S_z I_z \qquad \delta H = \sum_{j=x,y,z} \delta_j S_j$$



$$\omega_{S_0} = -\frac{A_\perp}{2} - \frac{A_\parallel}{4},$$

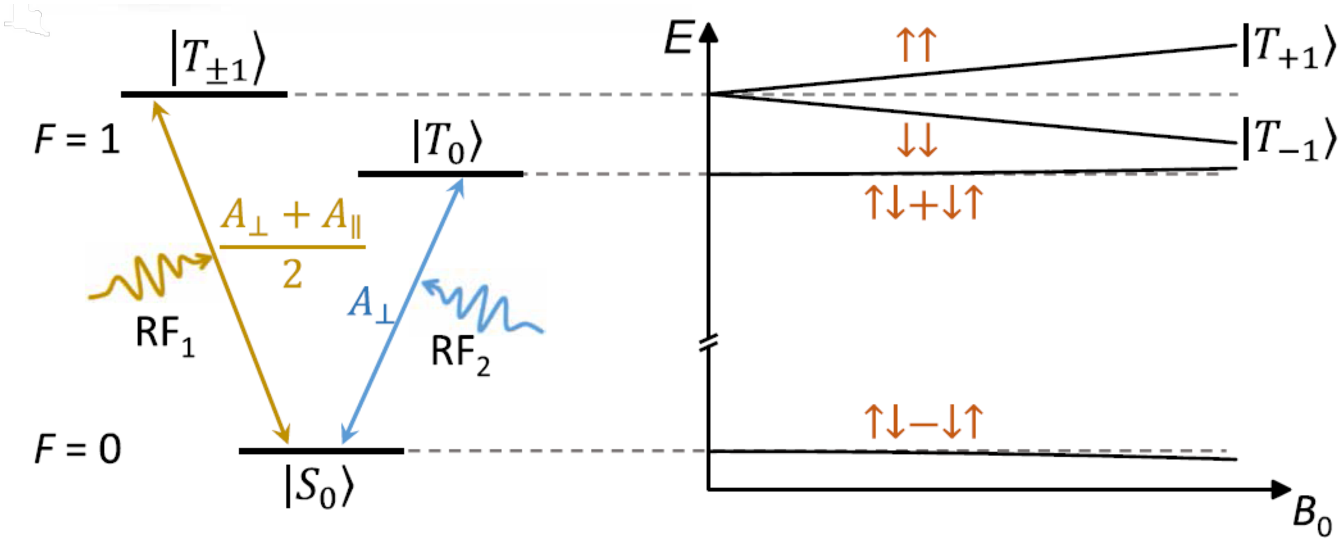
$$\omega_{T_0} = \frac{A_\perp}{2} - \frac{A_\parallel}{4},$$

$$\omega_{T_{\pm1}} = \frac{A_\parallel}{4}$$

$$\delta\omega_{S_0} \approx -\frac{\delta_x^{\,2} + \delta_y^{\,2}}{2(A_\parallel + A_\perp)} - \frac{\delta_z^{\,2}}{4A_\perp},$$

$$\delta\omega_{T_0} \approx -\frac{\delta_x^{\,2} + \delta_y^{\,2}}{2(A_\parallel - A_\perp)} + \frac{\delta_z^{\,2}}{4A_\perp},$$

$$\delta\omega_{T_{\pm1}} \approx \pm\frac{\delta_z}{2}$$

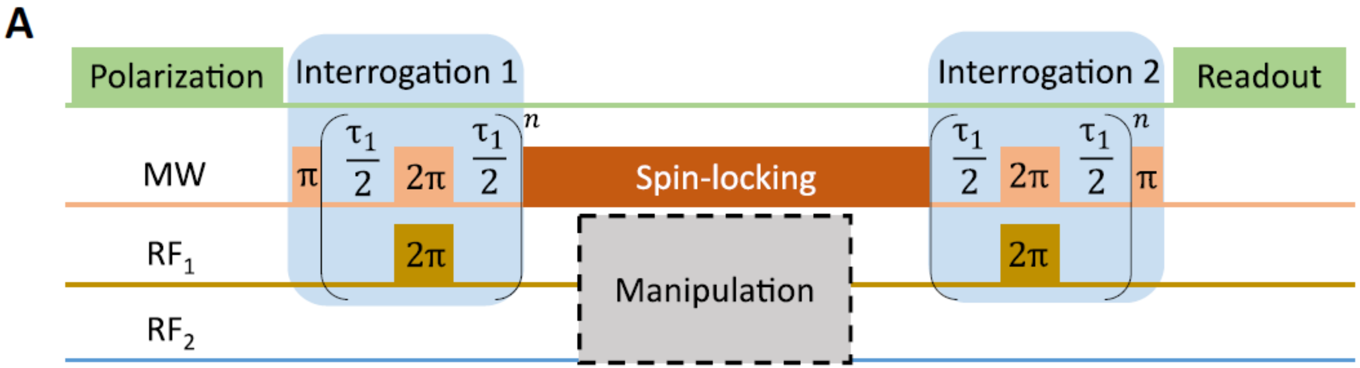


$A_\perp = 114$ MHz

$A_\parallel = 160$ MHz

Energy levels of the target spin

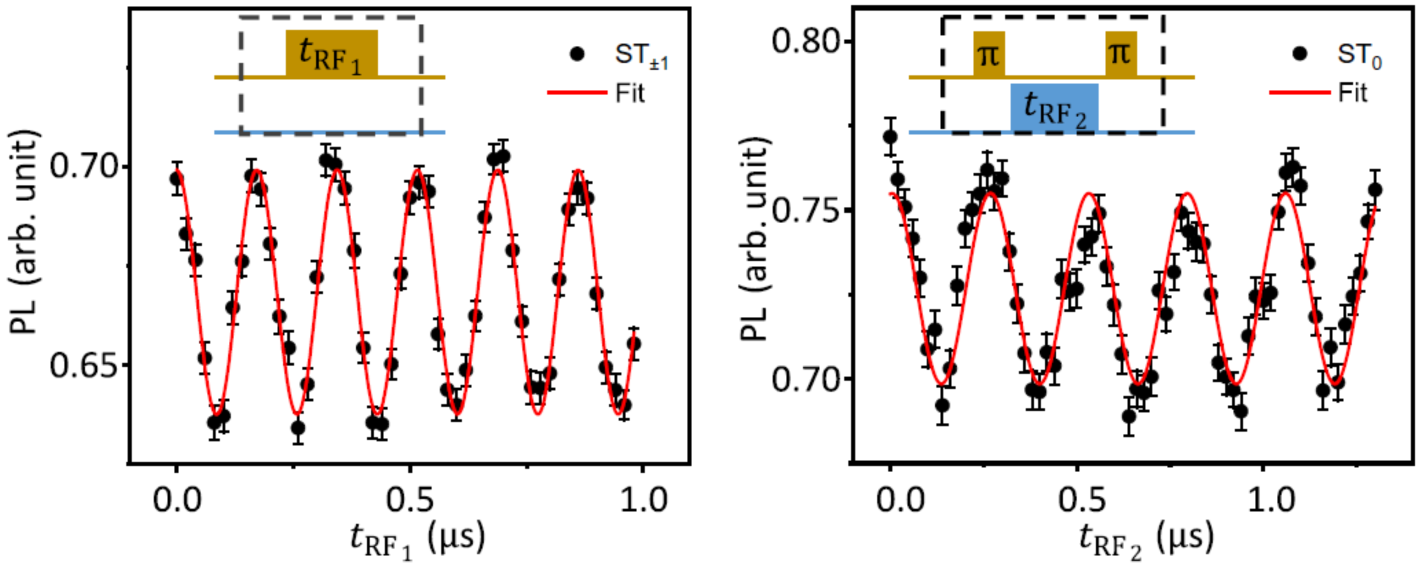
modified correlation detection protocol for zero-field EPR spectroscopy



$$S_{\mathrm{corr}} = \frac{1}{2}\left[1 + \langle \cos 2\varphi_1 \cos 2\varphi_2 \rangle\right]$$

$$|0\rangle \longrightarrow (|1\rangle + |-1\rangle)/\sqrt{2} \longrightarrow (e^{i\varphi_1}|1\rangle + e^{-i\varphi_1}|-1\rangle)/\sqrt{2}$$

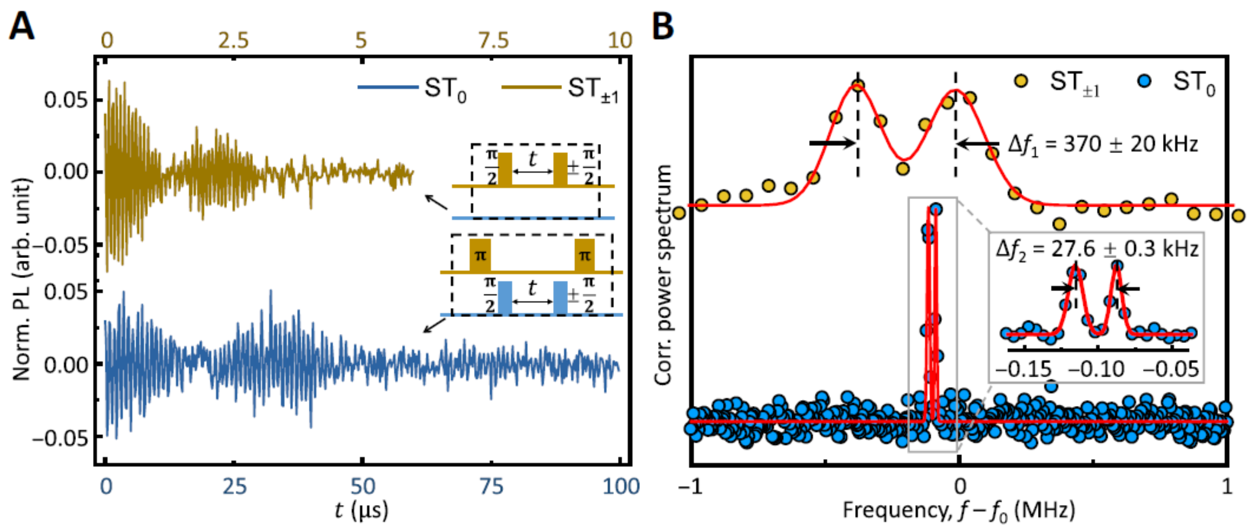
$$\cos\varphi_1 |\psi_+\rangle + i\sin\varphi_1 |\psi_-\rangle$$



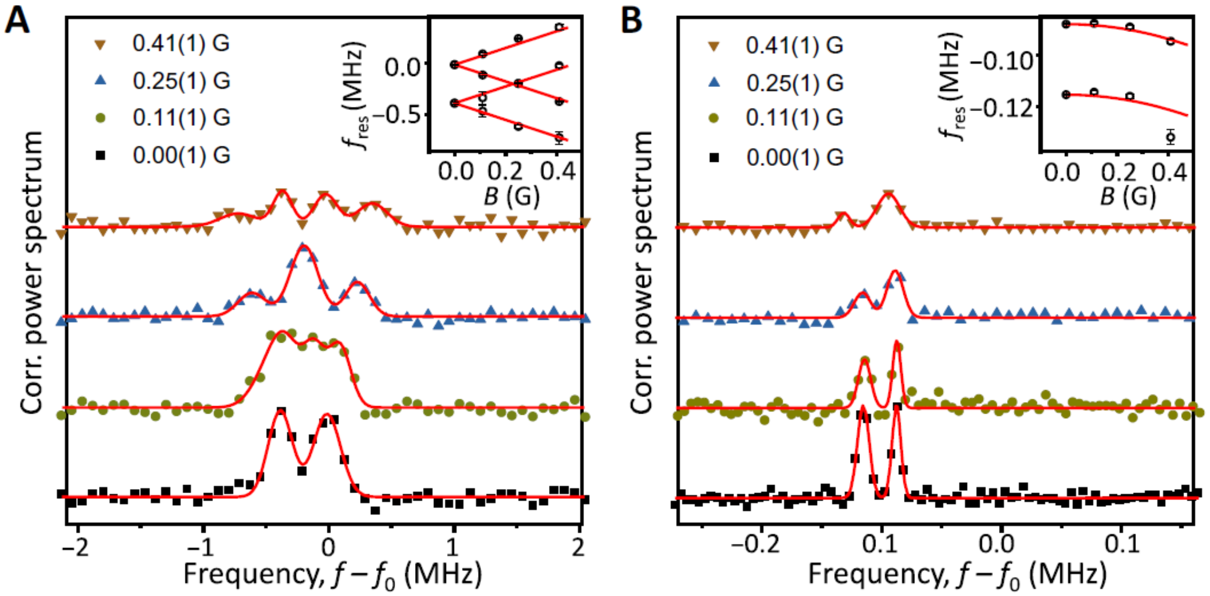
Rabi oscillations for the $ST_{\pm1}$ and ST_0 transitions.
f = 30 MHz, T_{1p} ~ 150 us

spin-locking relaxation time

High-resolution EPR spectroscopy of single P1 centers

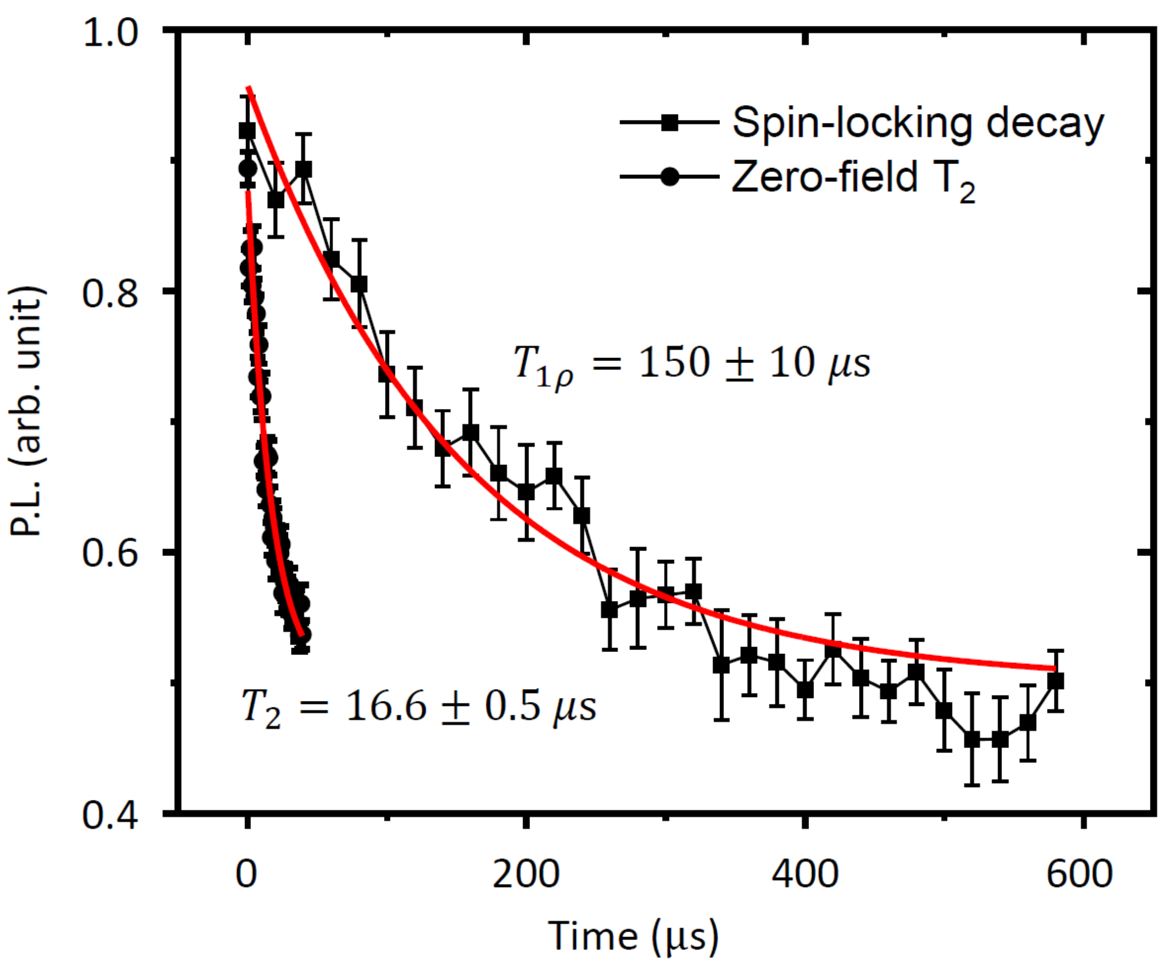


FWHM:
230±20 kHz and 260±20 kHz for $ST_{\pm1}$
11.6±0.6 kHz and 8.6±0.4 kHz for ST_0



Magnetic-field dependence
of the line shape
$ST_{\pm1}$ and ST_0

Coherence properties of the NV center



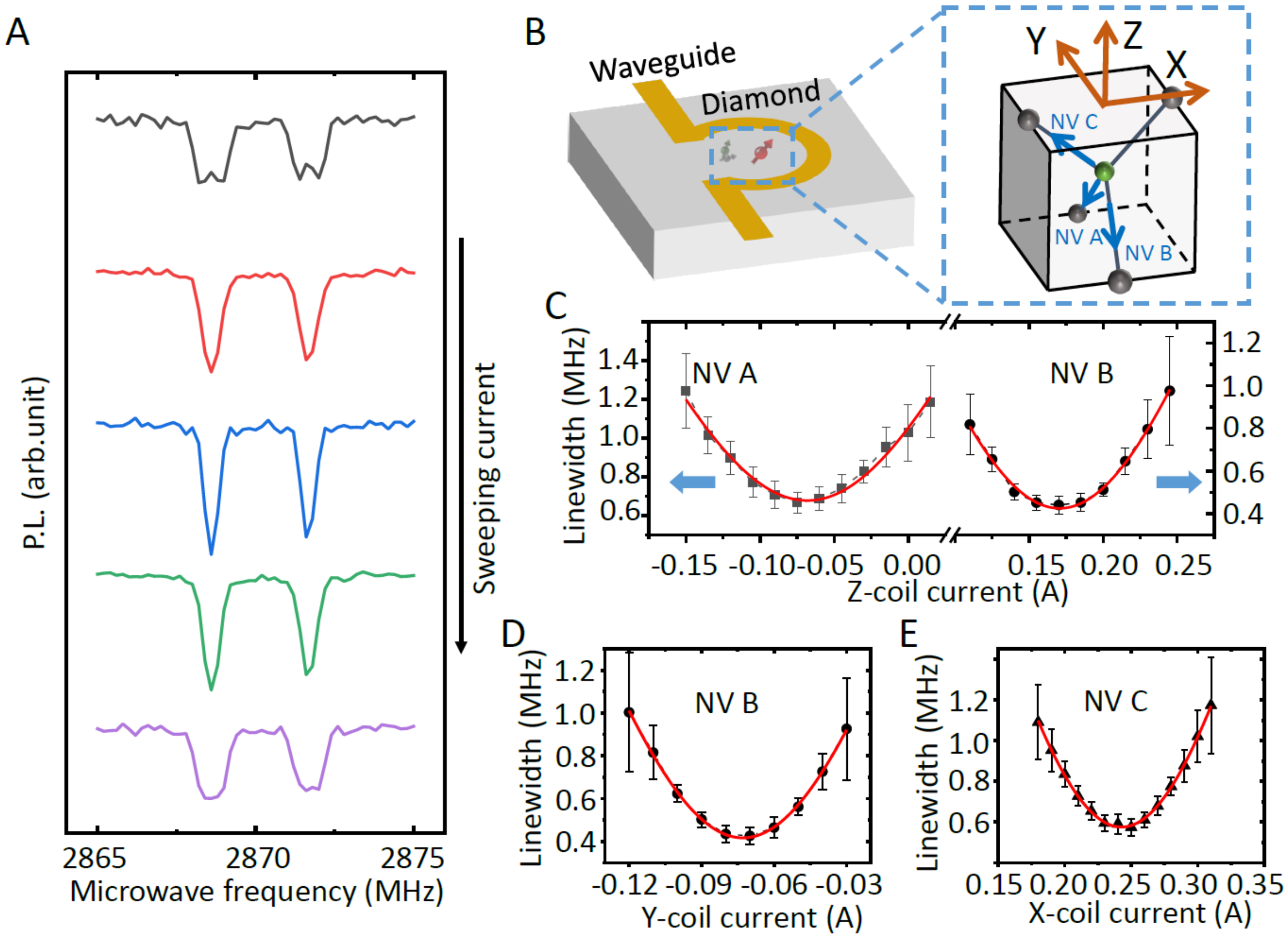
Magnetic-field dependence of the line shape

$$H_1 = \Omega_1 \cos\left[Dt + \frac{2\Omega_2}{\Omega_1}\sin\Omega_1 t\right] S_x$$

$$H_{\mathrm{II}} = e^{\mathrm{i}\frac{\Omega_1 t}{2}S_x} H_{\mathrm{I}}\, e^{-\mathrm{i}\frac{\Omega_1 t}{2}S_x} - \frac{\Omega_1}{2} S_x$$

$$= -\frac{\Omega_2}{2}(S_z^2 - S_y^2)$$

Schematics of the compensation process



Schematics of the compensation process