

Pressure Dependent Zero-Field Splitting in the Excited Triplet State of Diamond NV⁻ Centers

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The nitrogen-vacancy (NV) center in diamond is a versatile quantum sensor based on optically detected magnetic resonance (ODMR). In high-pressure physics, NV centers have been widely employed to visualize magnetic and pressure distributions at gigapascal (GPa) conditions [1-4]. Under 532 nm optical excitation, microwave resonance is detected as fluorescence dips originating from spin-dependent intersystem crossing between the triplet and singlet manifolds. At ambient pressure, the zero-field splitting (ZFS) is $D_{gs} \sim 2.87$ GHz in the ground state (GS) and $D_{es} \sim 1.42$ GHz in the excited state (ES), corresponding to transitions between $m_s = 0$ and $m_s = \pm 1$ sublevels. The pressure dependence of D_{gs} has been established to increase linearly with a slope of approximately 14.6 MHz/GPa [2,5,6], enabling quantitative pressure sensing. However, the pressure response of the excited-state ZFS D_{es} has so far been estimated only indirectly, from its temperature dependence [7].

In this work, we directly measure the pressure dependence of D_{es} up to the multi-GPa regime using diamond anvil cells (DAC) and evaluate the feasibility of excited-state ODMR for quantitative pressure sensing. In addition, the present study paves the way for future investigations of the ES lifetime's pressure dependence, aiming to clarify changes in the nonradiative intersystem crossing rate ($^3E \rightarrow ^1A_1$) under compression. Furthermore, our recent measurements revealed a substantial zero-field splitting in the excited-state ODMR spectra, which suggests the presence of significant local strain. Refining the Hamiltonian models for these high-pressure environments significantly improves the accuracy of our experimental data analysis. These results provide essential guidelines for optimizing ODMR measurements and extending NV-based quantum sensing to extreme-pressure environments.

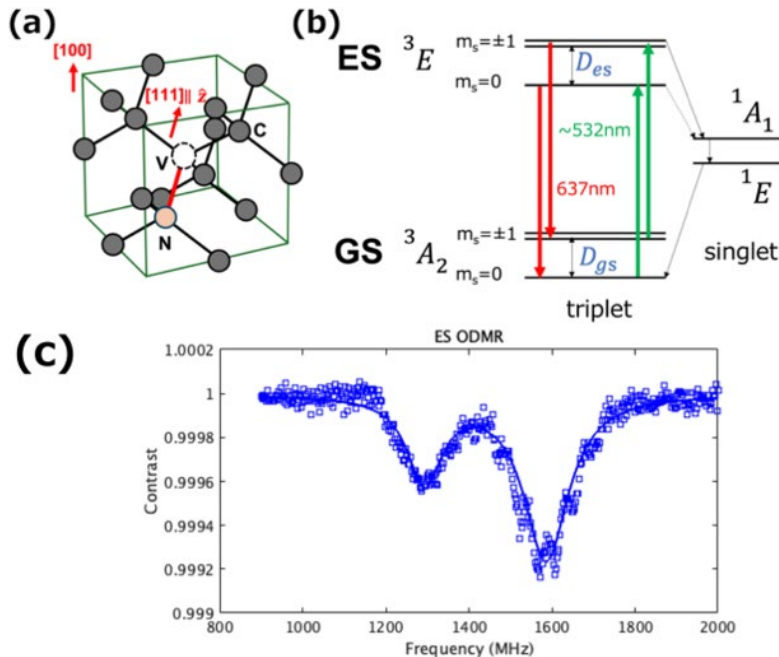


Fig (a): Schematic diagram of NV center (b): Optical transition system of NV center (c): ODMR fluorescence dip of ES and GS (3 GPa)

References

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