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# Ab-initio computation of EPR g tensor for point defects in solid state: results from the modern theory of orbital magnetization

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## Introduction

Electron paramagnetic resonance (EPR) is a very useful experimental technique to study point defects in solids. Its application holds particular significance in systems crucial to the development of quantum computing devices, notably within semiconductor frameworks. In this work, we utilize the modern theory of orbital magnetization [1] to compute the EPR g tensor, employing a “converse” approach implemented in the Quantum Espresso package [2]. Unlike the widely adopted linear-response method [3], the converse approach is nonperturbative. It may become advantageous in systems where spin-orbit coupling cannot be described as a perturbation or exhibits a large deviation from the free electron value [4].

## 1 Theoretical Background

The basic starting point is an independent particles Kohm-Sham Hamiltonian having the translation symmetry of the crystal but breaking Time-Reversal (TR) symmetry. Within the density-functional theory (DFT) and the all-electron (AE) formalism, the Hamiltonian in atomic units is:

$$H_{AE} = \frac{1}{2}[\mathbf{p} + \alpha A(\mathbf{r})]^2 + V(\mathbf{r}) + \frac{\alpha^2 g'}{8} \sigma \cdot [\nabla V(\mathbf{r}) \times \mathbf{p} + \alpha A(\mathbf{r})] \quad (1)$$

where  $A(\mathbf{r})$  is the symmetric gauge equal to  $\frac{1}{2}\mathbf{B} \times \mathbf{r}$ ,  $\alpha = \frac{1}{c}$  is the fine structure constant,  $g' = 2(g_e - 1)$  and  $\sigma$  are Pauli matrices. The spin-orbit coupling (SO) term is explicitly incorporated into the self-consistent field calculations while the spin other orbit (SOO) term, that in general has a small contribution to the g tensor, is neglected. The orbital magnetization for periodic systems and into norm-conserving pseudopotential framework is given by:

$$\mathbf{M} = \mathbf{M}_{bare} + \Delta\mathbf{M}_{NL} + \Delta\mathbf{M}_{para} + \Delta\mathbf{M}_{dia} \quad (2)$$

$$\mathbf{M}_{bare} = -\frac{\alpha N_c}{2N_k} Im \sum_{n,k} f_{n,k} \langle \partial_k u_{n,k} | \times (H_k + \epsilon_{n,k} - 2\epsilon_F) | \partial_k u_{n,k} \rangle \quad (3)$$

$$\Delta\mathbf{M}_{NL} = \frac{\alpha}{2} \sum_{\mathbf{R}} \langle (\mathbf{R} - \mathbf{r}) \times \frac{1}{i} [\mathbf{r} - \mathbf{R}, V_{\mathbf{R}}^{NL}] \rangle \quad (4)$$

$$\Delta\mathbf{M}_{para} = \frac{g'\alpha^3}{16} \sum_{\mathbf{R}} \langle (\mathbf{R} - \mathbf{r}) \times \frac{1}{i} [\mathbf{r} - \mathbf{R}, F_{\mathbf{R}}^{NL}] \rangle \quad (5)$$

$$\Delta\mathbf{M}_{dia} = \frac{g'\alpha^3}{16} \sum_{\mathbf{R}} \langle E_{\mathbf{R}}^{NL} \rangle \quad (6)$$

where  $H_k$  is the crystal Hamiltonian with  $\mathbf{B} = 0$ ,  $\epsilon_{n,k}$  and  $u_{n,k}$  are its eigenvalues and eigenvectors,  $\epsilon_F$  is the Fermi level,  $N_c$  is the number of cells and  $N_k$  is the number of k points. The  $|\partial_k u_{n,k}\rangle$  is  $\mathbf{k}$  the derivative of the Bloch wave function for  $n$  occupied states. The  $\langle \dots \rangle$  of Eq. 5-7 stands for  $\sum_{n,k} f_{n,k} \langle \psi_n | \dots | \psi_n \rangle$ , where  $V_{\mathbf{R}}^{NL}$  is the nonlocal pseudopotential in the separable Kleinmann-Bylander (KB) while  $F_{\mathbf{R}}^{NL}$  and  $E_{\mathbf{R}}^{NL}$ , are nonlocal operators accounting respectively the paramagnetic and diamagnetic contribution. Finally, from the orbital magnetization the

deviation of the g tensor  $\Delta g_{\mu\nu}$  from the free electron values ( $g_e = 2.002319$ ) is obtained by the following formula:

$$\Delta g_{\mu\nu} = -\frac{2}{\alpha S} e_{\mu} \cdot \mathbf{M}(e_{\nu}) \quad (7)$$

where  $\mu\nu$  are Cartesian directions of the magnetic field, and  $S$  is the total spin.

## 2 Application to point defects

Here following we present the EPR g tensor calculation for the substitutional Nitrogen defect in Silicon ( $N_{Si}$ ) in the off-center configuration (with  $C_{3v}$  symmetry) along one of the 111 crystal directions (Figure 1). This defect has recently garnered significant theoretical and experimental attention due to its potential role as a spin donor in silicon-based quantum devices [5, 6]. All ab-initio calculations were performed using the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [7], together with norm-conserving Trouiller-Martins pseudopotentials with GIPAW reconstruction and employing a plane wave basis set with a kinetic energy cutoff of 84 Ry. To model the off-center point defect we used a 512 atoms supercells in which a substitutional nitrogen atom is embedded within a silicon cell. We ensured convergence of the parameters by sampling k-points on a  $3 \times 3 \times 3$  mesh grid without considering symmetry, resulting in the integration of the Brillouin zone over a total of 54 k-points.

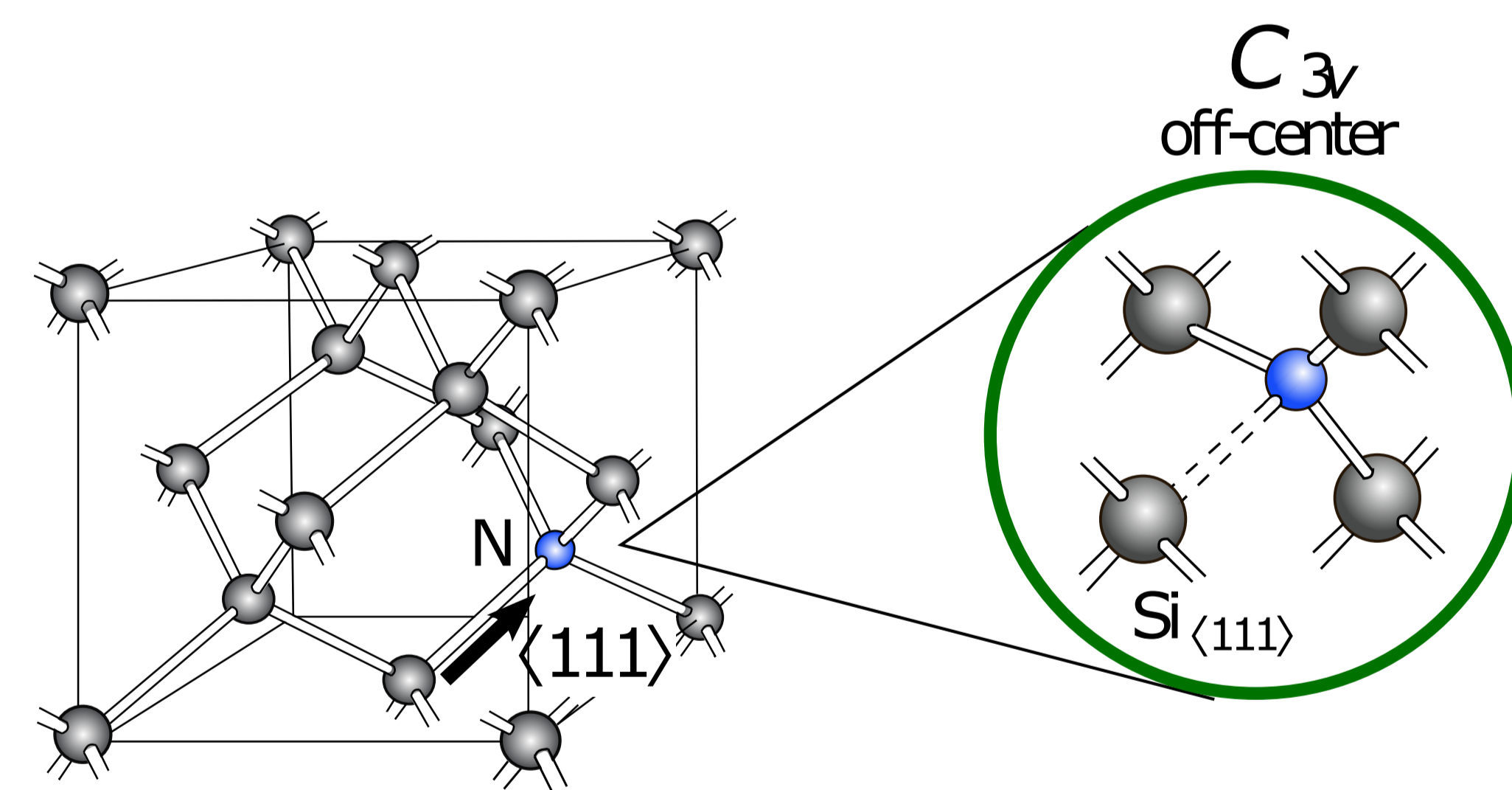


Figure 1: Graphic representation of the off-center configuration for  $N_{Si}$  defect

Ref.	$g_1$	$g_2$	$g_3$
This work	2.00209	2.00860	2.00860
Ref. [8]	2.00200	2.00890	2.00780
Exp. [5]	2.00219	2.00847	2.00847

Table 1: The EPR g tensor for the substitutional nitrogen point defect in silicon ( $N_{Si}$ ) calculated in this work compared with values obtained in Ref. [8] using a linear response approach. Results are compared to the available experimental data too.

## References

- [1] Ceresoli et al. *Phys. Rev. B*, 81:060409, Feb 2010.
- [2] Giannozzi et al. *Journal of Physics Condensed Matter*, 29(46), 2017.
- [3] Pickard et al. *Phys. Rev. Lett.*, 88:086403, Feb 2002.
- [4] Gerstmann et al. *Phys. Rev. B*, 81:195208, May 2010.
- [5] Belli et al. *Phys. Rev. B*, 89:115207, Mar 2014.
- [6] Belli et al. *Nanomaterials*, 14(1), 2024.
- [7] Perdew et al. *Phys. Rev. Lett.*, 77:3865–3868, Oct 1996.
- [8] Simha et al. *Nanomaterials*, 13(14), 2023.