Efficient Optimization of Interfacial Structures in Lattice-Mismatched Fe/MgO Interfaces Using Machine Learning and DFT Integration

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Magnetic Tunnel Junctions (MTJs) with Perpendicular Magnetic Anisotropy (PMA) are crucial for advanced memory technologies, particularly STT-MRAM [1]. The Fe/MgO interface is of significant interest due to its strong PMA and voltage-controlled magnetic anisotropy (VCMA) properties [2]. However, interfacial defects, exacerbated by the inherent lattice mismatch between Fe and MgO crystal structures, substantially impact device performance by altering magnetic properties and degrading crystal quality [3]. Recent studies have revealed that the Fe deposition method on MgO (001) significantly impacts crystal quality, with DC magnetron sputtering producing lower-quality crystals compared to other substrates [3]. Over-oxidation and under-oxidation in Fe_4N/MgO systems create various defects, while Density Functional Theory (DFT) calculations show that Frenkel and Schottky defects preferentially form at the MgO surface, affecting its structural properties [4], [5]. To address these challenges, this study employs the Global Optimization with First-principles Energy Expressions (GOFEE) method to explore interfacial defect structures in Fe/MgO (001) heterostructures [6]. DFT calculations are performed using the linear combination of atomic orbitals (LCAO) mode with the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional, k-point sampling density of 2.0 points per $Å^{-1}$ with gamma-point inclusion, and a double-zeta polarized (dzp) basis set. After 100 GOFEE iterations, two optimized Fe/MgO structures emerge with distinct characteristics. The first structure exhibits well-ordered body-centered cubic (bcc) Fe arrangement despite noticeable oxygen atom displacement in the MgO layer, while the second structure shows compromised bcc Fe crystallinity but maintains minimal oxygen shifts in the MgO layer. The correlation between oxygen atom positions and Fe crystal quality suggests that interfacial oxygen dynamics play a crucial role in determining the structural integrity of the Fe layer. These atomic-scale insights uncover subtle interfacial defect mechanisms that conventional methods might overlook, particularly the role of oxygen displacement in determining interface quality.

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