**QM/MM Investigations of Defects in MgO and Their Use as a Model System for High-*Tc* Superconductivity**

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The defect chemistry of magnesium oxide has been extensively studied for many years, with a myriad of applications in catalysis, optoelectronics and many other fields. Both vacancies and substitutionals have been shown to be effective in providing MgO with additional useful properties. Our work employs modern hybrid quantum mechanical – molecular mechanical (QM/MM) techniques, using the ChemShell code[1], to both reassess previously investigated defects and probe others that have been less researched.

Whereas a number of different transition metal ions have been investigated and widely applied as substitutionals in MgO, for example Ni and Co in catalytic systems, research into copper dopants in MgO is more sparse. Currently, the work on such systems is largely focused on nanoparticles, some uses in catalysis or the Jahn-Teller distortion induced by the dopant[2]. Our work focuses instead on the localisation and possible trapping of charge carriers within a (CuO6)10- unit. Whereas oxygen bound polarons have been observed with a number of doped metal oxides[3], currently, there is no evidence for the influence that copper has, in this regard. Hybrid-DFT within QM/MM has been utilised to track the migration of such holes, with further electromagnetic resonance (EPR) calculations allowing for direct comparison to experimental work. Our work will be used to guide future investigations of hole states and bipolaron formation in superconducting cuprate materials.

Additionally, the optical excitations of F-centres within MgO have been investigated and compared to previous theory of their excited states. It has been well reported that excited states of electrons within oxygen vacancies in MgO are hydrogenic in character with distortions from the crystal field environment[4,5]. Through use of time-dependent DFT (TDDFT) within a QM/MM scheme, these characteristics are reinvestigated.

References:

[1] Y. Lu et al., J. Chem. Theory Comput. 15, 1317 (2019).  
[2] M. J. Riley, C. J. Noble, and P. L. W. Tregenna-Piggott, The Journal of Chemical Physics 130, 104708 (2009)  
[3] O. F. Schirmer, physica status solidi c 4, 1179 (2007)  
[4] G. H. Rosenblatt et al., Phys. Rev. B 39, 10309 (1989)  
[5] C. Jun et al., Eur. Phys. J. B 9, 593 (1999)

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