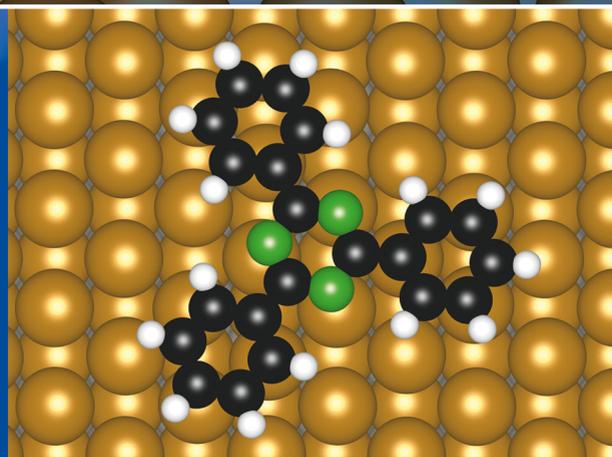
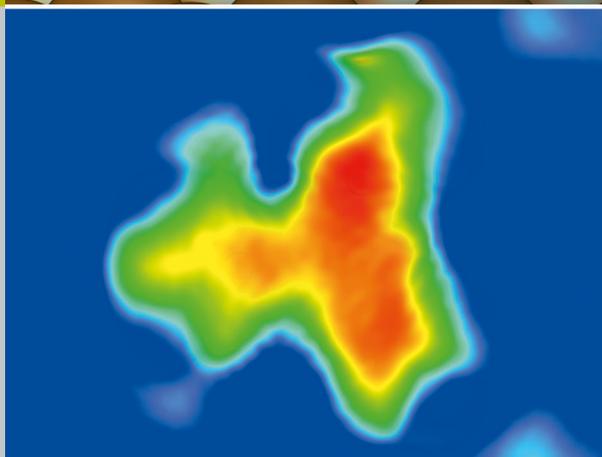
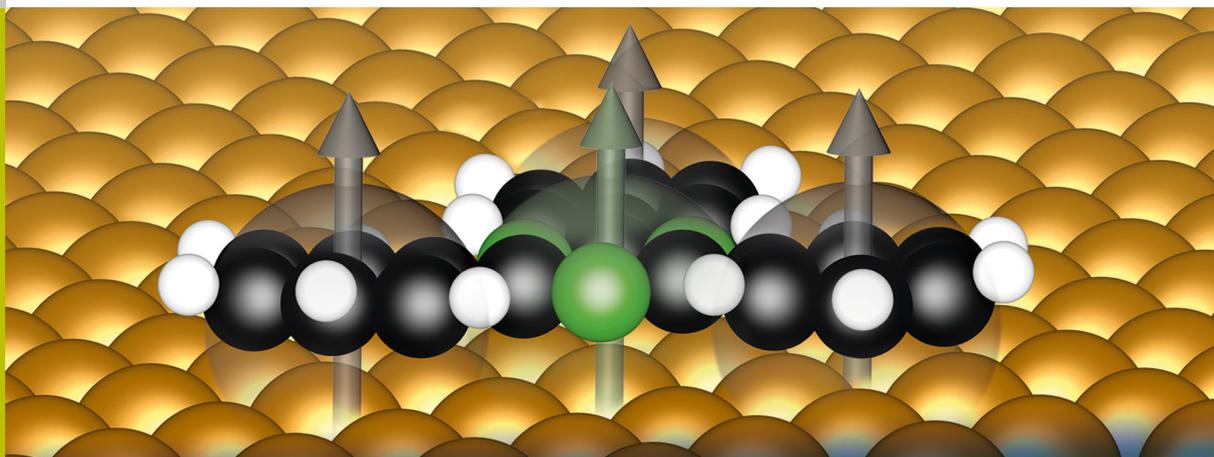


Ab initio investigation of hybrid molecular-metallic interfaces as a tool to design surface magnetic properties for molecular spintronics

Rico Friedrich



Schlüsseltechnologien /
Key Technologies
Band / Volume 138
ISBN 978-3-95806-194-1

Forschungszentrum Jülich GmbH
Peter Grünberg Institute (PGI)
Quantum Theory of Materials (PGI-1 / IAS-1)

Ab initio investigation of hybrid molecular-metallic interfaces as a tool to design surface magnetic properties for molecular spintronics

Rico Friedrich

Schriften des Forschungszentrums Jülich
Reihe Schlüsseltechnologien / Key Technologies

Band / Volume 138

ISSN 1866-1807

ISBN 978-3-95806-194-1

1. Introduction	13
1. Basics	19
2. Survey of Electronic Structure Theory	21
2.1. The Many-body Problem in Electronic Structure Theory	21
2.1.1. The Many-body Hamiltonian	21
2.1.2. The Born-Oppenheimer Approximation	23
2.1.3. Approaches to Electronic Structure	25
2.2. Density Functional Theory	27
2.2.1. The Thomas-Fermi Approach	27
2.2.2. The Hohenberg-Kohn Theorems	28
2.2.3. The Kohn-Sham Equations	29
2.2.4. Approaches to the Exchange-correlation Functional	32
2.2.5. Generalization to Spin-polarized Systems	39
2.2.6. Extensions and Recent Developments	40
2.3. Practical Considerations for Actual Calculations	41
2.3.1. Basis Sets	41
2.3.2. The Pseudopotential Approach	44
2.3.3. The Projector Augmented-wave Method	48
2.3.4. The Green's Function Approach of Korringa, Kohn and Rostoker	53
2.3.5. Connection of DFT to Electronic Transport	58
3. Magnetic Interactions	63
3.1. Origin of Magnetism and Theoretical Models	63
3.1.1. Origin of Magnetism	63
3.1.2. Exchange Coupling of Two Spins	65
3.1.3. Heisenberg Model	66
3.1.4. Calculation of the Exchange Coupling Constant	67
3.1.5. Hubbard Model	69
3.2. Exchange Mechanisms	69
3.2.1. Direct Exchange	69
3.2.2. Superexchange	71
3.2.3. Double Exchange	73

3.2.4. RKKY Interaction	74
3.2.5. Higher Order Exchange	76
3.2.6. Magnetic Anisotropy and Antisymmetric Exchange	76
4. Introduction to Molecular Spintronics	79
4.1. Preliminaries from Molecular Electronics	79
4.1.1. Short Overview of Molecular Electronics	79
4.1.2. Physisorption and Chemisorption	81
4.2. Origin and Categorization of Spintronics	84
4.2.1. The Giant Magnetoresistance Effect	84
4.2.2. Categorization of Spintronics	86
4.2.3. Origin of Organic and Molecular Spintronics	88
4.3. Overview of Important Developments in Molecular Spintronics	91
4.3.1. Spectroscopic Investigations	91
4.3.2. Transport Studies Through Very Thin Organic Layers	92
4.3.3. Investigations of Single Molecule-Surface Hybrid Systems	94
4.3.4. Spintronic Investigations Using Single-Molecule Magnets	97
4.3.5. Spin Crossover Molecules for Molecular Spintronics	98
4.3.6. Organic Magnetoresistance	99
4.3.7. Spin and Chirality	99
4.4. Tuning Surface Magnetic Exchange by Molecular Adsorption	100
4.4.1. Inter-layer Magnetic Softening Effect	101
4.4.2. In-plane Magnetic Hardening Effect	102
4.4.3. Deduced Questions	104
II. Results	107
5. Chemical Tuning of Magnetic Hardening	109
5.1. Presentation of the Molecular Species	110
5.2. Adsorption Geometries and Structural Properties	112
5.3. Magnetic Moments and Exchange Coupling Constants	115
5.4. Spin-polarized Projected Density of States	121
5.5. Magnetic Anisotropy Energies	123
5.6. Multi-scale Monte Carlo Simulations	124
5.7. Conclusions	126
6. Molecular Induced Skyhook Effect	129
6.1. Presentation of the Molecular Species	130

6.2. Adsorption Geometries and Structural Properties	131
6.3. Magnetic Moments and Exchange Coupling Constants	133
6.4. Spin-polarized Projected Density of States	140
6.5. Conclusions	143
7. Magnetic Exchange Below 2D Materials	145
7.1. Presentation of the 2D Materials	146
7.2. Adsorption Geometries and Structural Properties	148
7.3. Magnetic Moments and Exchange Coupling Constants	151
7.4. Spin-polarized Projected Density of States	155
7.5. Conclusions	156
8. Formation of Different Magnetic Units	159
8.1. Presentation of the TPT Molecule	160
8.2. Adsorption Geometries and Structural Properties	161
8.3. Simulated STM Images and Spin-polarized Projected Density of States	166
8.4. Magnetic Moments and Exchange Coupling Constants	171
8.5. Conclusions	175
9. Molecular Tuning of the Rashba Effect	177
Introduction to the Rashba Effect	178
9.1. Presentation of the Molecular Species	182
9.2. Presentation of the Clean Surface Alloy	183
9.3. Adsorption Geometries and Structural Properties	188
9.4. Charge Density Difference and Electrostatic Potential	190
9.5. Band structures and Rashba-split Surface States	193
9.6. Conclusions	203
10. Summary and Outlook	205
III. Appendix	209
A. Computational Details	211
A.1. The Vienna <i>Ab Initio</i> Simulation Package	211
A.2. The Spin Polarized Relativistic Tight-binding Korringa-Kohn-Rostoker Program .	211
A.3. Visualization for Electronic and STructural Analysis	212
A.4. Common Computational Details	212

A.5. Small π -electron Systems on 1 ML Fe/W(110)	213
A.5.1. Convergence Tests	214
A.5.2. Energy-dependent exchange coupling constant of 1 ML Fe/W(110)	215
A.5.3. Distance Dependence of the Exchange Coupling Constants	215
A.5.4. Small π -electron Systems with Third Period Elements on 1 ML Fe/W(110)	216
A.6. Dioxan and Dioxin on 1 and 2 ML Fe/W(110)	221
A.6.1. SP-PDOS for the Molecular-induced Geometries	221
A.7. Gr and hBN on Co(111)	222
A.7.1. Convergence Test	222
A.7.2. Results Obtained with the PBE Functional	223
A.8. TPT on 2 ML Fe/W(110)	224
A.8.1. SP-PDOS of the Fe <i>d</i> -states for TPT on 2 ML Fe/W(110)	224
A.9. Ammonia (NH ₃) and Borane (BH ₃) on BiAg ₂ /Ag(111)	224
A.9.1. Convergence Tests	225
A.9.2. Effective masses of the surface states for NH ₃ and BH ₃ on BiAg ₂ /Ag(111)	226
A.10. TPT on 2 ML Co/Cu(111)	226
A.11. Graphene on 1 ML Fe/Co on Cu(111)	226
B. TPT on 2 ML Co/Cu(111)	229
B.1. Adsorption Geometries and Structural Properties	229
B.2. Simulated STM Images and Spin-polarized Projected Density of States	232
B.3. Magnetic Moments and Exchange Coupling Constants	236
C. Graphene on 1 ML Fe/Co on Cu(111)	239
C.1. Adsorption Geometries	239
C.2. Spin-polarized Density of States and Coupling Constants of the Clean Surfaces	239
C.3. Spin-polarized Density of States and Coupling Constants of the Hybrid Systems	241
List of Figures	245
List of Tables	249
List of Abbreviations	251
Bibliography	255
Publications	275

**Schlüsseltechnologien /
Key Technologies
Band / Volume 138
ISBN 978-3-95806-194-1**

