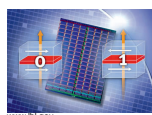
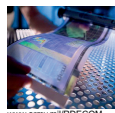


Motivation

Merging spintronics and organic electronics

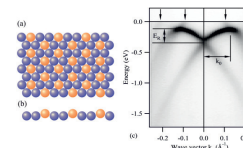
- organic molecules become building blocks of more and more electronic devices: low cost, easy to process, etc.
- the electronic properties of the organic molecules can easily be tuned, e.g., by the choice of the central atom of Pc molecules
- spintronics: spin dependent transport phenomena are used as an additional information channel besides the regular electric currents to construct, e.g. smaller devices with larger storage capacities
- combination of both fields paves the way to future high performance electronic devices



Giant Spin Orbit Splitting

Splitting due to an in-plane gradient

- in surface alloys such as BiAg₂, PbAg₂, and BiCu₂, the larger atomic species (Bi, Pb, etc.) buckles out of the surface plane
- due to the buckling, these alloys exhibit an in-plane potential gradient that leads to a spin splitting of the surface bands ("giant spin splitting") apart from and even higher than the Rashba type spin splitting (which is due to a potential gradient in z-direction found, eg., for Au(111), Bi(111))
- giant spin splitting is about 3 times larger than the Rashba type splitting observed for Bi(111), about 10 times larger than for Au(111)
- the surface state of the original metal (Ag, Cu, ...) vanishes upon formation of the giant spin split alloy surface state



Material	E_s (meV)	k_s (Å ⁻¹)	α_s (eV/Å)	Reference
BiAg ₂ /BiAg ₂ heterostructure	<1	0.028	0.07	[1]
Ag(111) surface state	<0.2	0.004	0.03	[2,3]
Au(111) surface state	2.1	0.012	0.33	[4,5]
Bi(111) surface state	-14	-0.05	-0.56	[6]
Bi/Ag(111) surface alloy	200	0.13	3.09	This work

E_s : Rashba energy of split states
 k_s : wave number offset
 α_s : Rashba parameter

C.R. Ast et al., PRL **98**, 186807(2007)

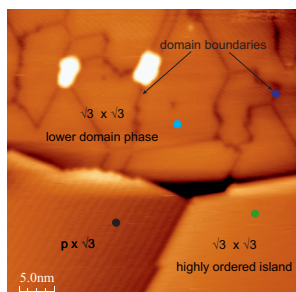
BiAg₂ Surface Alloy / BiAg₂ Mixed Structures

Preparation

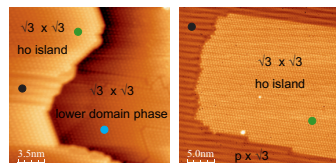
- evaporation of Bi from a Knudsen cell onto a pre-heated Ag(111) single crystal
- post annealing step to about 100°C
- formation of BiAg₂ surface alloy checked via LEED

General sample structure

- sample covered with $\sqrt{3} \times \sqrt{3}$ highly ordered BiAg₂ islands / domains
- lower BiAg₂ $\sqrt{3} \times \sqrt{3}$ ordered areas exhibit numerous domain boundaries
- slight excess of Bi leads to a gradual dealloying of the $\sqrt{3} \times \sqrt{3}$ highly ordered BiAg₂ islands into $p \times \sqrt{3}$ structures
- $p \times \sqrt{3}$ islands: $\sqrt{3}$ periodicity in Ag[112] direction (lower, striped appearance)



$\sqrt{3} \times \sqrt{3}$ BiAg₂ surface alloy



left: V_{sample} = 0.7 V, I_r = 30 pA, T = 80 K
 right: V_{sample} = 0.1 V, I_r = 55 pA, T = 80 K



left: V_{sample} = 0.7 V, I_r = 30 pA, T = 80 K
 center: V_{sample} = 1.0 V, I_r = 100 pA, T = 80 K
 right: V_{sample} = 0.6 V, I_r = 55 pA, T = 80 K

- upon landing, the Bi atoms embed into the Ag(111) substrate
- to relieve surface stress this dilute surface alloy becomes a more and more dense $\sqrt{3} \times \sqrt{3}$ structure
- displaced Ag atoms diffuse around the surface and form highly ordered $\sqrt{3} \times \sqrt{3}$ islands with incoming Bi atoms
- after the entire surface is covered with the BiAg₂ surface alloy further Bi leads to a dealloying of the sample by transforming $\sqrt{3} \times \sqrt{3}$ ordered islands into $p \times \sqrt{3}$ ordered islands

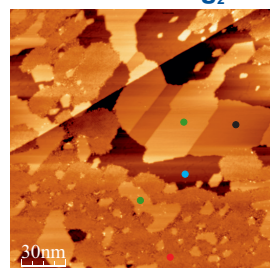
Literature

C. Kato, Y. Aoki, and H. Hirayama, PRB **82**, 165407 (2010)

K.H.L. Zhang et al., PRB **83**, 235418 (2011)

C. R. Ast et al., PRB **75**, 201401(R) (2007)

PTCDA on BiAg₂



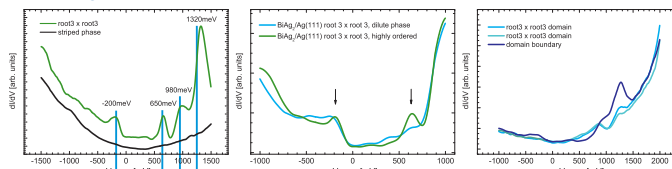
V_{sample} = 1.2 V, I_r = 10 pA, T = 80 K

General sample structure

- at submonolayer coverage PTCDA seems to grow exclusively on lower BiAg₂ covered areas
- as on several other metallic substrates, PTCDA forms ordered island where the molecules assume a herringbone like arrangement
- PTCDA forms large islands surrounding the highly ordered $\sqrt{3} \times \sqrt{3}$ islands
- several domain boundaries can be found within the molecular layers
- while some PTCDA covered areas exhibit almost perfect order, large areas exhibit a certain disorder in their herringbone arrangement

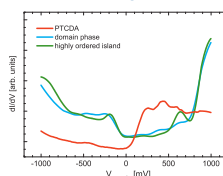
Spectroscopy

Pure BiAg₂ structures



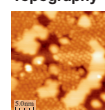
- the BiAg₂ spectra exhibit a peak which can be assigned to the giant spin split alloy surface state as well as several peaks corresponding to empty states
- all empty state peaks are in good agreement with DFT calculations (G. Bihlmayer et al., PRB **75**, 195414 (2007)) and earlier STS data (C.R. Ast et al., PRB **75**, 201401(R) (2007))
- slight differences in the spectra of BiAg₂ islands and lower structures are observed
- domain boundaries within the lower level BiAg₂ structures show a different spectroscopic behavior than the inner part of the domains (C.R. Ast et al., PRB **75**, 201401(R) (2007))
- spectra of the $p \times \sqrt{3}$ structure remains almost featureless within an energy range of ± 1.5 V around the Fermi level

PTCDA on BiAg₂

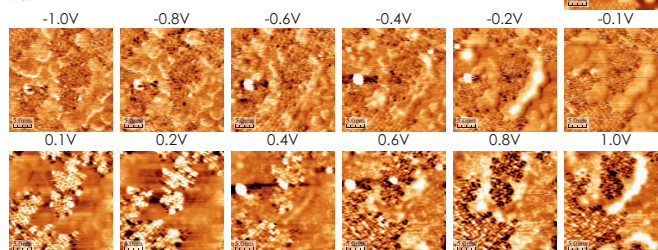


- no features observed at the energy of the BiAg₂ surface state in spectra taken on PTCDA layers
- pronounced onset of PTCDA molecular levels close to E_F for empty states

Topography



dI/dV



I_r = 100 pA, T = 80 K, Topography image recorded at V_{sample} = -0.8 V

Outlook: Interaction of PTCDA with the Bi-Ag surface structures

- Is the disorder induced by the substrate?
- Is the interaction with the substrate stronger at the $\sqrt{3} \times \sqrt{3}$ domain boundaries?
- Do the molecules contribute to relieve/ build up surface stress?
- Is there any interaction between the molecular overlayer and the giant spin orbit split surface state?

→ Laterally resolved STS in combination with ARPES measurements

V_{sample} = 1 V, I_r = 100 pA, T = 80 K



Funding

Studienstiftung
des deutschen Volkes

CENIDE
CENTER FOR NANOTECHNOLOGY
DUISBURG-ESSEN

DFG Deutsche
Forschungsgemeinschaft

SFB616 „Energy Dissipation at Surfaces“



Special thanks to GxSM and WSxM

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