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Hg-based epitaxial materials for topological insulators

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# UNIVERSIDAD AUTONOMA DE MADRID UNIVERSITARIA DE CANTO BLANCO MADRID 28049 SPAIN

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Topological insulators (TI) stand as a new class of materials in which spin-orbit interaction produces topologically nontrivial insulating bulk electronic phases and conducting material boundaries. These boundary regions show complex spin textures and have properties that are unlike any other known 2D electronic systems with foreseen applications ranging from topological superconductors and spintronics to quantum computation. Recent trends in TI consist in searching for new candidate 2D materials like heavy element (high Z) magnetic TI. In particular, diluted magnetic semiconductors such as Hg(1-x)M(x)VI, where M is a transition metal (e.g., Mn, Fe, Cr) and VI is a Group VI element (e.g., Te, Se), are of special interest due to their variable band-gap and excellent electronic-transport and optical properties. In this study, TI structures were made by sandwiching a thin layer of Hg-based material between Cd(Zn)Te substrates and cover layer of CdTe. Several geometries and compositions were produced with controllable topological structure using both vapor-phase epitaxy and liquid-phase epitaxy, along with associated microstructural characterization. Samples were provided to the Air Force Research Laboratory for investigation of properties.					
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## Project title: <u>Hg-based epitaxial materials for topological insulators</u>

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- 2. Summary of the project results
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## 1. Brief project description

Topological insulators (TI) stand as a new class of materials in which spin-orbit interaction produces topologically nontrivial insulating bulk electronic phases and conducting material boundaries. These boundary regions show complex spin textures and have properties that are unlike any other known two dimensional electronic systems with foreseen applications ranging from topological superconductors and spintronics to quantum computation (e.g. see C.L.Kane and J.E.Moore "Topological Insulators" Physics World (2011) 24, 32). The topological insulator states in 2D and 3D materials were predicted theoretically in 2005 and in 2007 the first key experiment in this field was the observation of the 2D quantum spin Hall effect in a quantum well structure made by sandwiching a thin layer of HgTe between layers of Hg<sub>1-x</sub>Cd<sub>x</sub>Te.

Recent trends in TI consist in searching for new candidate 2D materials like heavy element (high Z) magnetic TI, for example, tetradymite semiconductors  $Bi_2Te_3$ ,  $Bi_2Se_3$ , and  $Sb_2Te_3$  which form magnetically ordered insulators when doped with transition metal elements Cr or Fe (Rui Yu et al. "Quantized Anomalous Hall Effect in

Magnetic Topological Insulators" Science 2 July 2010: 61-64) or diluted magnetic semiconductors (DMS) Hg<sub>1-x</sub>Mn<sub>x</sub>Te (H. Buhmann "Towards the quantum anomalous Hall effect in HgMnTe" MAR12-2011-001728, MAR12 Meeting of the American Physical Society). Among these compounds, Hg-based DMS materials, in particular those of the type Hg<sub>1-x</sub>M<sub>x</sub>VI (M–Mn, Fe, Cr; VI-Te, Se), could be of special interest for TI because their variable band-gap and excellent electronic-transport and optical properties are driven by an external and internal magnetic fields via a strong spin exchange interaction of band carriers with localized magnetic ions (J.K. Furdyna "Diluted magnetic semiconductors" J. Appl. Phys. 64 (1988) 29).

Similarly to other TI candidate materials, an important functional drawback of DMS was represented by the fact that until recently the transition temperature into spinglass phase (T<sub>g</sub>) in these materials was relatively low, making difficult their practical use (for example, T<sub>g</sub>~20K in Hg<sub>1-x</sub>Mn<sub>x</sub>Te at x=0.45). Lately, our studies of several Hgbased DMS such as Hg<sub>1-x</sub>Cr<sub>x</sub>Se and Hg<sub>1-x-y</sub>Cr<sub>x</sub>Mn<sub>y</sub>Se have surprisingly shown that they exhibit spin-glass transitions at considerably higher temperatures T<sub>g</sub>≥100K (S.Yu.Paranchych et al. "Effect of structural and compositional inhomogeneities on spin-glass transition in Hg<sub>1-x-y</sub>Cr<sub>x</sub>Mn<sub>y</sub>Se crystals." J. Crystal Growth 262 (2004) 403-407 and references therein). This fact could potentially open the way to realize the TI materials operating at high temperate and, thereby, make them practically important for numerous applications.

<u>Scientific and technological objectives:</u> The fabrication of Hg-based epitaxial materials of several geometries and compositions with controllable topological structure for exploring their feasibility for TI.

#### 2. Summary of the project results

To attain the project objectives, the TI structures were made by sandwiching a thin layer of Hg-based material between CdTe substrate and the cover layer of CdTe. There were used the experimental setups similar to those we have early reported, in particular for the CdTe growth (N.V. Sochinskii et al. "Growth and characterization of CdTe:Ge:Yb" J. Crystal Growth 310, (2008) 2076–2079), the Hg<sub>1-x</sub>Mn<sub>x</sub>Te LPE (N.V.Sochinskii et al. "Structural properties of Hg<sub>1-x</sub>Mn<sub>x</sub>Te layers grown on CdTe substrates by liquid phase epitaxy" Semicond. Sci. Technol. 11 (1996) 542-547), and the Hg<sub>1-x</sub>Cd<sub>x</sub>Te VPE (N.V. Sochinskii et al. "Vapor phase epitaxy of Hg<sub>1-x</sub>Cd<sub>x</sub>Te on CdTe heteroepitaxial substrates" J. Crystal Growth 149 (1995) 35-44).

According to the project workplan, the experimental work was developed in two work packages such as (I) Technology development of Hg-based epitaxial materials of several geometries and compositions with controllable topological structure and (II) Structural characterization of Hg-based epitaxial materials for TI. In particular, the work consisted in

- Setting up of technological facilities required for the project development: thermal evaporator for VPE experiments based on rapid thermal annealing (RTA) approach, substrate holders, crucibles;

- Preparation and fabrication of starting materials: CdTe substrates, VPE sources of several materials and compositions (HgTe, Hg<sub>1-x</sub>Mn<sub>x</sub>Te, Hg<sub>1-x-y</sub>Cr<sub>x</sub>Mn<sub>y</sub>Se);

- Epitaxial growth of Hg-based epitaxial materials of several geometries and compositions;

- Experimental study of the VPE and LPE setups and time-temperature conditions;

The most important project results are as follows:

- the fabrication of the experimental samples of Hg-based epitaxial heterostructures of about 100 sq. cm. total area;

- the lot of preselected samples was delivered;

- the samples were characterized by SEM and EDAX techniques;
- based on the experimental findings, further research was proposed (Annex I).

#### 3. Characterization data

3.1 LPE growth

The LPE growth was carried out on Cd(Zn)Te substrates from Hg-rich growth solutions in the temperature range of 360-400 °C. Using our LPE growth approach, it was possible to fabricate sandwiched CdTe/HgCd(Mn)Te/CdZnTe heterostructures with a relatively thick (several microns) HgCd(Mn)Te intermediate layer (Figure 1) as well as several intermediate Hg-based layers of different compositions (Figure 2). At the same time, up to moment, we had no success in the fabrication of LPE heterostrutures with very thin (below 1  $\mu$ m) intermediate layer due to planarity problems (Figure 3).

A – CdTe cover layer



# B - HgCd(Mn)Te internal layer



# C - CdZnTe substrate



Figure 1. SEM images of a typical CdTe/HgCd(Mn)Te/CdZnTe LPE heterostructure with the EDAX composition data in indicated areas.



Figure 2. SEM images of a CdTe/HgCd(Mn)Te/CdZnTe LPE heterostructure with several intermediate layers.



Figure 3. SEM images of a CdTe/HgCd(Mn)Te/CdZnTe LPE heterostructure with thin intermediate layer.

3.2 VPE growth

The VPE growth was carried out on Cd(Zn)Te substrates from Hg(Cd)Te sources by rapid vapor deposition (RVD) technique in the temperature range of 600-700°C. Using our VPE growth approach, it was possible to fabricate sandwiched CdTe/Hg(Cd)Te/CdZnTe heterostructures with the thin (below 10-100 nm) Hg(Cd)Te intermediate layers of very good planarity and different compositions (Figure 4).

The usage of the catalyst assisted VPE growth resulted, at the moment, in hardly controlled dendrite-like growth that needs further improvements (Figure 5).



Figure 4. SEM images of a CdTe/HgCdTe/CdZnTe LPE heterostructure with thin intermediate layers.

# A – dendrite shape



B – general surface view



Figure 5. SEM images of HgCdTe/CdZnTe VPE heterostructure with a dendrite-like layer.

#### 4. Delivered samples

Ι	Sample fabrication method:				
	LPE (liquid phase epitaxy) from Hg-rich growth solutions				
	Sample name	Sample specification			
	(number and				
	fabrication date)				
	1-20/09/13	CdTe/HgCdMnTe/CdZnTe heterostructure (the LPE of			
	2-15/10/13	HgCdMnTe 5-40 um enilaver followed by the LPE of CdTe			
	3-29/10/13	rigeawinite 5-46 µm epilayer followed by the EFE of eare			
	4-08/11/13	5-10 μm epilayer)			
	5-13/11/13				
	6-13/11/13				
II	Sample fabrication method: VPE (vapor phase epitaxy)				
	Sample name	Sample specification			
	7-11/11/13	CdTe/HgTe/CdZnTe heterostructure (the VPE of several nm			
		thick HgTe epilayer on CdZnTe bulk substrate followed by			
		the VPE of about 100 nm thick CdTe epilayer; the CdTe			
		epilayer surface were polished with diamond solution)			
	8-18/11/13	CdTe/HgTe/CdZnTe heterostructure (similar to previous			
		sample 7; as-grown surface – no polishing)			
	9-19/11/13	HgCdTe/HgTe/CdZnTe heterostructure (commercial single-			
	10-04/11/13	crystalline (111) oriented substrate; polished surface)			
	11-18/11/13				
	12-14/11/13				
III	Sample fabrication method: VBG (vertical Bridgman growth)				
	Sample name	Sample specification			
	13- $Hg_{1-x}Cr_xSe$	x=0.1; wafer with several single crystalline blocks; cut of the			
		crystal perpendicularly to the growth axis; mechanically			
		polished			

#### 5. Conclusions

We have demonstrated the fabrication of Hg-based epitaxial materials of several geometries and compositions with controllable topological structure by the LPE and VPE methods. The lot of selected samples has been delivered for investigating their properties. Based on the gathered experience, it has been developed the proposal for further technological and physical research needed for exploring the feasibility of Hg-based heterostructures for TI purposes.