Peculiarity of Superconducting and Galvanomagnetic Properties of Bicrystals of 3D Topological Insulator Bi_{1-x} Sb_x $(0,07 \leq x \leq 0,2)$

F. M, MUNTYANU[1, 2], A. GILEWSKI[2], A. J. ZALESKI[3], T. PALEWSKI[2], V.MUNTEANU[1]
and V. CHISTOL[4]

[1] *Institute of Electronic Engineering and Industrial Technologies, ASM, Chisinau, 2028, Moldova*
[2] *International Laboratory of High Magnetic Fields and Low Temperatures, Wroclaw, 53421, Poland*
[3] *Institute of Low Temperatures and Structural Research, Polish Academy of Sciences, Wroclaw, 50950, Poland*
[4] *Technical University of Moldova, Chisinau, 2004, Moldova*
muntean_teodor@yahoo.com

Abstract — **We present the results of investigation of magnetic properties and high-field (up to 40 T) galvanomagnetic phenomena in bicrystals of 3D topological insulator Bi_{1-x} Sb_x $(0.07 < x < 0.22)$ with nano-width interfaces (~100 nm). It has been found that the onset of superconducting transition in these bicrystals is almost 36K, the semiconductor–semimetal transition is induced in crystallites, adjacent and central layers of interfaces at different values of quantum magnetic field, the superconductivity and ferromagnetism coexist in some crystallite interfaces.**

Index Terms — **bismuth-antimony alloys, semiconductor–semimetal transition, superconductivity, ferromagnetism**

I. INTRODUCTION

According to recent studies [1-3], the single crystals of Bi_{1-x} Sb_x alloys in the semiconducting regime $(0.07 < x < 0.22)$ are three- dimensional 3D topological insulator (TI) that belongs to a non-trivial Z_2 topological class. The 3D TI is a novel quantum state of matter [4] and is insulating in the bulk but have gapless edge/surface states. The results of angle-resolved photoemission spectroscopy show that the surface states of Bi_{1-x} Sb_x $(0.07 < x < 0.22)$ alloys are metallic. Since the surface states surround the sample, the quantum transport phenomena in magnetic fields became very attractive [5] for studying the metallic surface states in the Bi_{1-x} Sb_x $(0.07 < x < 0.22)$ topological insulators. In bicrystals of these alloys, consisting of two single crystalline blocks (3D TI) and the superconducting nano-width crystallite interface (CI), one of the external surfaces is replaced with the CI which in turn is a complex systems composed of a solitary central part (the thickness about 60nm) and two similar adjacent layers (~ 20nm) on both sides of it. In spate of small size, the CIs play a significant role in electronic transport [6] and may change qualitatively the distinctive behavior of topological insulators.

The evolution of the band structure of the bulk Bi_{1-x} Sb_x alloys with increasing Sb concentration *x* is highlighted by decreases up to zero (x~0,07) of the overlap between electron minima at L-points and hole maximum at T- point of the Brillouin zone; band inversion at L points (x~ 0,04); semimetal-to-semiconductor transition at x~0,07 and vice versa at x~0,22 . If $x > 0,22$ the Bi_{1-x} Sb_x alloys are semimetals as a result of overlapping of L_a and H bands specific to antimony [5]. The surfaces band structure of semiconducting Bi_{1-x} Sb_x alloys has been studied for a while and is clearly identified only for (111) face; it consists [2,3] of single electron pocket surround Γ point and six elliptical hole pockets centered along the six line connecting Γ and M points of the surface Brillouin zone. The band structure of CIs of Bi and Bi_{1-x} Sb_x alloys is investigated in [6] and is revealed that Fermi surface consisting of layer components is similar to those in bulk specimens, but the shape, elongation, and volume of isoenergetic surfaces undergo essential changes.

Here, we report the results of study of magnetic properties and galvanomagnetic phenomena in high magnetic fields having as a objective the elucidation of the role played by superconducting nano-width interfaces in the interaction between Dirac fermions in a 3D TI.

IV. EXPERIMENTAL PROCEDURE

The bicrystals of Bi_{1-x} Sb_x $(0.07 < x < 0.22)$ alloys were obtained by the zone recrystallization method using the double seed technique. The width of crystallite interfaces (~100nm) was estimated by means of scanning electron microscopy (SEM) and by the magnetic field value at which quantum oscillations become observable [6]. The samples for measurements were prepared in the form of parallelepipeds ($1x2x4mm^3$). The composition of the samples were controlled by a SEM equipped with Oxford and PV 9800 energy-dispersive X-ray (EDX) analyzers and by the methods for optical emission spectrometry using a Jobin-Yvon spectrometer JY 38 S. The magnetic impurity concentrations were evaluated in some samples being found : Cr ~ (27-47) ppm, Ni ~ (145- 496) ppm, and Fe < 0,02 ppm. The magnetic properties of Bi_{1-x} Sb_x

bicrystals was studied in the temperature range (1.6 - 30)K and fields up to 7T using a Quantum Design SQUID magnetometer and a Physical Property Measurement System. The measurements were carried out in the Institute of Low Temperatures and Structure Research of the Polish Academy of Sciences in Wroclaw. The transport phenomena were studied in stationary up to 18T and pulse magnetic fields up to 40T in the International Laboratory of High Magnetic Fields and Low Temperatures (Wroclaw, Poland).

III. RESULTS AND DISCUSSION

The semiconducting $Bi_{1-x}Sb_x$ ($0.07 < x < 0.22$) alloys are 3D TI [1,2] - material with a bulk electronic excitation gap, generated by the spin-orbit interaction, which allows novel electronic properties both for single crystalline samples and bicrystals. Below, we list three of these manifestations connected with peculiarities of the bulk and interfaces energy band structure.

The first case relates to the semiconductor-semimetal transition in high magnetic fields. Fig.1 presents the field dependences of magnetoresistance $\Delta\rho/\rho$ in bicrystals at $B \parallel C_3$ (in crystallites).

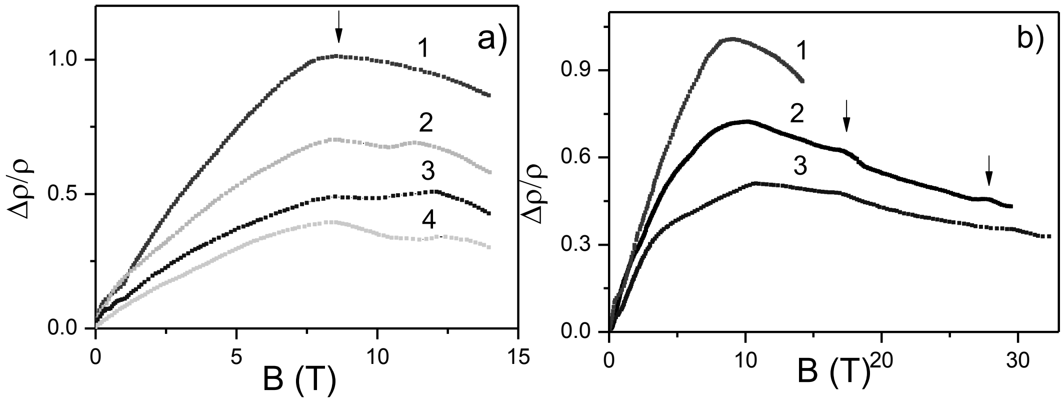

Fig.1. Semiconductor - semimetal transition in bicrystals of 3D topological insulator $Bi_{1-x}Sb_x$ ($0.07 < x < 0.22$) at 4,2K. (a) Stationary magnetic fields. 1. single crystalline sample, x=0.08; 2,3,4- bicrystals, x= 0.08; 2.$\Theta_1 = 4^o$, $\Theta_2=2^o$; 3. $\Theta_1 = 1^o$, $\Theta_2=4^o$; 4.$\Theta_1 = 9^o$, $\Theta_2=2^o$, (b) Pulse magnetic fields, 1. single crystalline sample, x=0.08; 2,3- bicrystals, 2. x= 0.08, $\Theta_1=4^o$, $\Theta_2=2^o$; 3. x= 0.09, 2. $\Theta_1 = 12^o$, $\Theta_2=2^o$.

As shown, the quantum oscillations $\Delta\rho/\rho$ are displayed up to 0.2T. In magnetic fields B > 0,2T charge carriers are in the ultra quantum limit (UQL), where the electrons (or holes) occupy the zeroth Landau level and the band edge displacement $\Delta\varepsilon$ takes place, depending on the ratio of spin $\Delta\varepsilon_s$ and orbital $\Delta\varepsilon_0$ level splitting: $\Delta\varepsilon= 1/2h\omega(1- \Delta\varepsilon_s / \Delta\varepsilon_0)$. In single crystalline samples of $Bi_{1-x}Sb_x$ ($0.08 < x < 0.12$) alloys the band edge displacement in high magnetic field is essential and the semiconductor-semimetal transition occurs [7] at B ~ (8 – 9) T (see the maximum of curve 1 in Fig.1a,b). In bicrystals this maximum is detected at the same field values (Fig.1a, curve 2-4 and Fig1b, curve 1), which means that the semiconductor - semimetal transition is induced in crystallites. In addition to maximum at 9T, against the monotonic decrease of $\Delta\rho/\rho$, in high fields were revealed two peaks related to CI (in single crystalline samples they do not appear), more exactly with the semiconductor-semimetal transition in adjacent and central layers. We remark that the transitions were observed at different values of quantum magnetic field; hence, the ratio of spin and orbital level splitting $\Delta\varepsilon s / \Delta\varepsilon_0$ differ considerably [6] in crystallites, adjacent and central layers of bicrystals. This implies a significant increase of electronic excitation gap due to growth of spin-orbit interaction at interfaces and necessitates the existence of gapless electronic states on the bicrystal boundary.

The second case relates to observation of superconductivity in 3D TI. One or two superconducting phases are identified (see Fig.2a,b) on temperature dependences of the magnetic moment m(*T*) and resistivity ρ(T) [9] of bicrystals of $Bi_{1-x}Sb_x$ ($0.07 < x < 0.22$) alloys. The first superconducting phase (is shown independently or together with the second phase), depending on sample composition, is characterized by transition temperature variations within the range (3.7 - 4.6) K. On the other hand, Tc of the second phase changes considerably (8.3 - 36) K. The presence of superconducting transitions on dependencies *m*(*T*) and ρ(T) is remarkable because crystallites of Bi_{1-x} - Sb_x ($x \le 0.2$) alloys, as well as of Bi, are not superconductors. According to [8], these transitions are connected with two unknown superconducting phases located at CIs, namely in adjacent and central layers of bicrystals. In both phases, we observed that upper critical field H_{c2} varies approximately linearly with temperature suggesting that the anisotropy of H_{c2} is temperature independent. The estimated values of ($- dHc2 / dT$) for superconducting phases lie in the range 0.5 - 3.5 kOe/ K. Large magnetic hysteresis loops characteristic of type-II superconductors were observed with lower critical field H_{c1}~ (100–130) Oe. The hysteresis loops are almost symmetric (Fig.2b), exhibits nearly reversible behavior (the irreversibility field ~ (1.5– 4 kOe), their form does not change essentially with temperature and sometimes does not display an initial vortex penetration peak. These features suggest that interaction between Dirac fermions in a topological insulator is coherently controlled by superconducting phases.

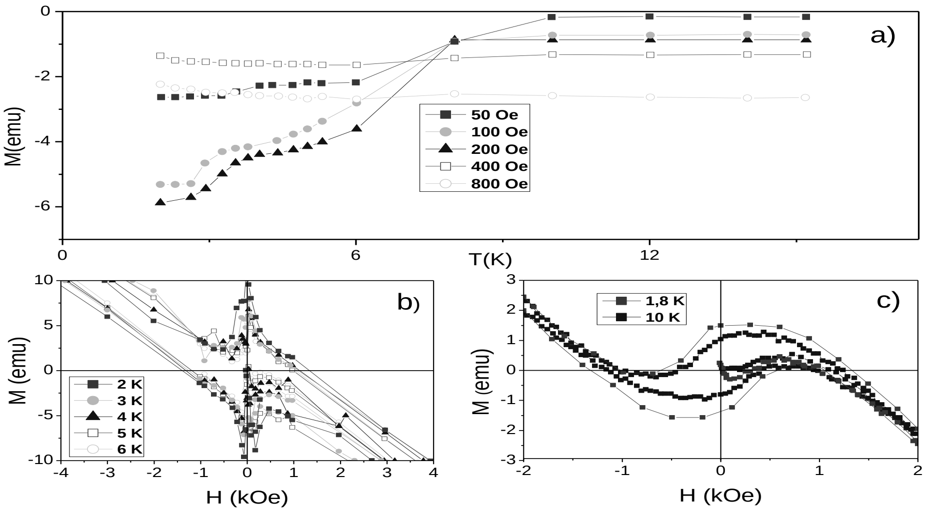

Fig.2. (a) Temperature dependences of the magnetic moment of bicrystal of $Bi_{1-x}Sb_x$ (x =0.15), 2.$\Theta_1 = 4^o$, $\Theta_2=2^o$; (b) Magnetic hysteresis loops of $Bi_{1-x}Sb_x$ (x =0.15), 2.$\Theta_1 = 4^o$, $\Theta_2=2^o$; (c) Magnetic hysteresis loops of $Bi_{1-x}Sb_x$ (x =0.15), 2.$\Theta_1 = 12^o$, $\Theta_2=5^o$.

The third case relates to coexistence of superconductivity and feromagnetism in $Bi_{1-x}Sb_x$ (0.08 < x <0.12) bicrystals. As we seen from the Fig. 2c in some of our bicrystals on paramagnetic dependences of m(T), the ferromagnetic-like hysteresis loops was found (calculated contributions in m(T) of the residual magnetic impurities are one order smaller than the experimentally recorded values) in the entire temperature interval under study despite the diamagnetic response at T<5K. A similar situation was reported for another semimetal–graphite. No noticeable change in the form and width of hysteresis loops on temperature was detected. The observed occurrence of superconductivity and ferromagnetism appears to be a consequence of the increase in the carrier density at the nano-width CIs and changes in the carrier pockets topology [6]. In this regard, the CIs (in comparison with bulk single crystals) as topologically ordered systems are intrinsically robust against local sources of decoherence; therefore, the increase in volume and change of the shape and elongation of isoenergetic surfaces of charge carriers specifies the common electronic origin of superconductivity and ferromagnetism, which (together with the updating of the phonon spectra) favor the spin independent coupling or lead to the magnetic ground state at the wide structural disorder and a plenty of topological defects.

CONCLUSION

We studied the magnetic and galvanomagnetic properties at low temperatures (1,6 – 30)K and high magnetic fields (up to 40T) in bicrystals of 3D topological insulator $Bi_{1-x}Sb_x$ (0.07 < x <0.22) and it was found: (i) The ratio of spin and orbital level splitting differs considerably in crystallites, adjacent and central layers of bicrystals; (ii) The superconducting phases are identified on temperature dependences of magnetic moment of bicrystals, whereas the crystallites are not superconductors; (iii) The coexistence of superconductivity and ferromagnetism has been observed in some bicrystals.

REFERENCES

[1] L. Fu and C. L. Kane, „Topological insulators with inversion symmetry". Phys. Rev. B **76**, 045302 (2007).

[2] J. C. Y. Teo, Liang Fu, and C. L. Kane, „Surface states and topological invariants in three-dimensional topological insulators: Application to Bi1 − xSbx," Phys. Rev. B. **78**, 045426 (2008).

[3] D.Hsieh, D.Qian, L.Wray, Y.Xia, Y.S.Hor, R.J.Cava and M.Z.Hasan, „A topological Dirac insulator in a quantum spin Hall phase", Nature (London) **452**, 970 (2008).

[4] Hai-Zhou Lu, Wen-Yu Shan, Wang Yao, Qian Niu, and Shun-Qing Shen, „Massive Dirac fermions and spin physics in an ultrathin film of topological insulator", Phys. Rev. B.81, 115407 (2010).

[5] F.M.Muntyanu, V.G.Kistol, and I.A.Popov, „Quantum oscillations of transport phenomena of H-holes in $Bi_{1-x}Sb_x$ ($x \geq 0.25$)", Phys. Stat. Sol. (b) 148, K37 (1988).

[6] F. M. Muntyanu, A. Gilewski, K. Nenkov, A. Zaleski and V. Chistol, „Fermi-surface rearrangement in Bi bicrystals with twisting superconducting crystallite interfaces" Phys. Rev. B **76**, 014532 (2007).

[7] D.V.Gitsu, F.M.Muntyanu, M.I.Onu, and A.I.Russu, „Magnetoresistance anisotropy of $Bi_{0,91}Sb_{0,09}$ alloy in strong magnetic field", Phys. Stat. Sol. (b) **93,** K127 (1979).

[8] F. M. Muntyanu, A. Gilewski, K. Nenkov, J. Warchulsca, and A. Zaleski, „Experimental magnetization evidence for two superconducting phases in Bi bicrystals with large crystallite disorientation angles" Phys. Rev. B **73**, 132507 (2006).

[9] F. M. Muntyanu, A. Gilewski, K. Nenkov, A. Zaleski, T. Palewski and V. Chistol, „High-field peculiarities of galvanomagnetic quantum oscillations in Bi bicrystals with nano-width superconducting crystallite interfaces", Solid State Comm (2013), http://dx.doi.org/10.1016/j.ssc.2012.12.018i.