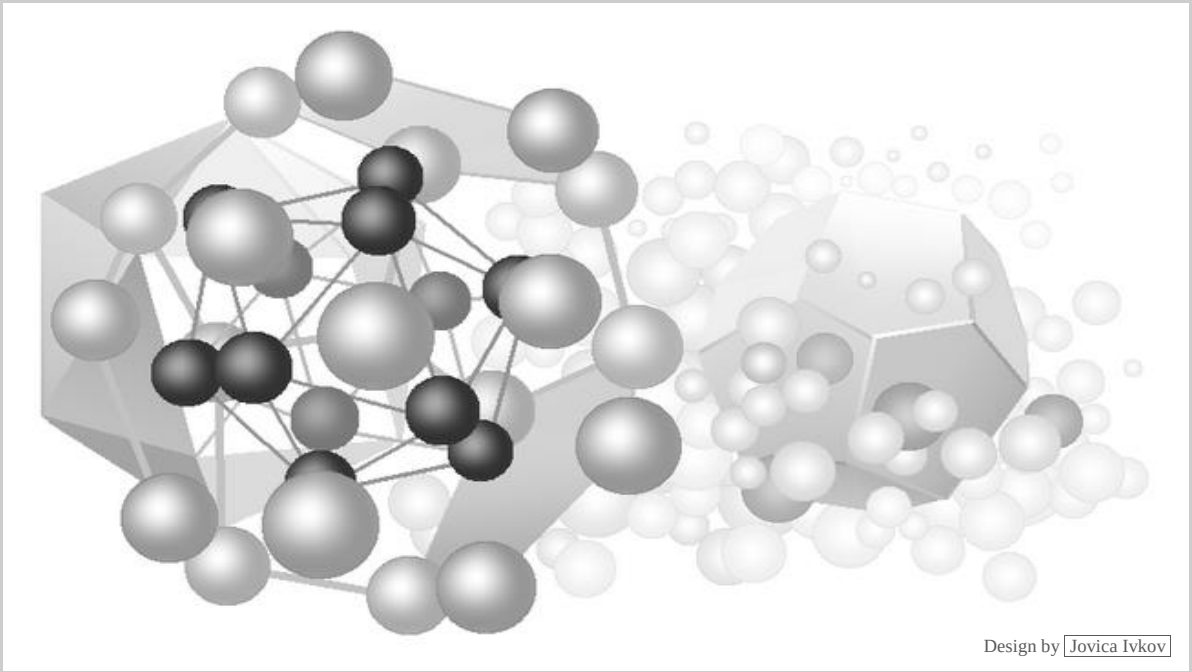
S y m p o s i u m

in memory of

Jovica Ivkov



Design by Jovica Ivkov

February 24, 2017
Institute of Physics, Zagreb, Croatia

Program and Abstracts

Symposium "*One-day scientific meeting in memory of Jovica Ivkov***"** is organized by the Institute of Physics, Zagreb, Croatia

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Jovica Ivkov: Beauty of Science, Education and Art

The scientific community has recently lost an excellent experimental condensed matter physicist. Jovica Ivkov passed away quietly as a result of cancer, on June 26, 2016 in Zagreb. He made important contributions to physics, including metallic glasses, complex metallic alloys, amorphous thin films and high temperature superconductors.

Jovica Ivkov was born on July 12, 1953, in Belgrade. He finished elementary school in Belgrade and secondary school in Zagreb. Jovica Ivkov showed excellence in physics already in high school. He successfully participated in national physics competitions. During physics studies at the Faculty of Science, University of Zagreb, he was one of the best students (among others are Bojana Hamzić, Željko Marohnić, Mladen Prester, Silvia Tomić …). He started scientific research during his diploma work by joining the investigation of metallic glasses within the group *Physics of metals II* (in Croatian *Fizika metala II*) at the Institute of Physics of the University of Zagreb. Due to his conceptual knowledge of physics, analytical thinking and experimental skills he was from the beginning an equal member of the group. Thus when finalizing his diploma work in 1978, he was already a coauthor of six scientific papers. It is interesting to note that his first paper published in 1977 was already based on measurements of Hall effect. After graduation, he was employed as a physics teacher in a high school in Jastrebarsko (1979 - 1982). This left a trace on his scientific output in this period despite his efforts to continue with scientific activities.

From 1982 Jovica Ivkov was continuously employed at the Institute of Physics of the University of Zagreb. This was just the beginning of his very successful career characterized by his exceptional modesty and originality (during which he influenced many prominent scientists at home and abroad). From the beginning Jovica Ivkov dominated research of galvanometric properties (especially Hall effect) for which he independently constructed measurement devices and protocols. One has to bear in mind that in the 80-ties the scientific equipment at the Institute of Physics of the University of Zagreb was quite scarce. Therefore, Jovica Ivkov performed majority of his measurements for his master's thesis in solid state physics on the NMR magnet of the Institutes of Biology, Physics, University of Zagreb, when it was available, very often on weekends. Only in the 90-ties did Jovica Ivkov completely build his setup which he improved for years. Simultaneously, Jovica Ivkov has taught classes at Faculty of Science, University of Zagreb (Exercises in Basic Physics I - IV). He received his MSc degree in physics from the University of Zagreb, 1987, and started his PhD studies in solid state physics which he finished in 1991. Despite numerous and diverse responsibilities, he published twenty remarkable scientific papers in the first ten years at the Institute of Physics of the University of Zagreb. Standing out among those contributions, was the work on the connection between electronic structure obtained by studying Hall coefficient (R_H) and physical properties of two most important systems among metallic glasses: transition metal – metalloid (TL-M, eg. $Fe_{80}B_{20}$) and early transition metal – late transition metal (TE – TL, eg. $Zr_{1-x}Ni_x$) (contribution of E. Babić). In the case of TL-M metallic glasses (Ni based alloys) he got the correct numbers of conducting electrons per atom in a broad region of the concentration of the metalloid. This enabled him to describe the change of the electronic structure with the change of concentration and nature of metalloid atoms. Furthermore, using the number of electrons per atom that he determined, he managed to demonstrate that Ziman – Faber model describes fairly well transport

properties of those metallic glasses. Further investigation on magnetic TL-M metallic glasses (Fe, Co, Ni-BSi) showed that mechanism of anomalous Hall effect changes with the increase of the concentration of magnetic component (Fe, Co), and that R_H is still closely linked with its electronic structure. In the case of TE-TL metallic glasses he was the first to point to the connection between the change of the sign of R_H and the gap between d-band of TE and TL in those metallic glasses (one of his most cited papers). He also studied the very interesting effect of hydrogen doping on R_H and electronic structure in TE-TL metallic glasses (contribution of M. Novak). In 2013 he again started investigating R_H in TE-TL metallic glasses (contribution of R. Ristić), this time aiming at establishing the connection between their electronic structure and the ease of glass formation.

From the 90-ties, Jovica Ivkov simultaneously investigated high temperature superconductors (especially in his focus were monocrystals of $Bi_2Sr_2CaCu_2O_x$, contribution of D. Babić) and amorphous films of normal metals (eg. Al, Cu) and refractor metals in collaboration with scientists from Institute of Ruđer Bošković (contributions of N. Radić and M. Sorić). He continued investigation of amorphous films until the end of his scientific career. From the late 90-ties he started challenging research on complex metallic alloys (that includes quasicrystals, a new state of condensed matter) in the framework of the group of Ana Smontara (contributions of J. Dolinšek, Z. Jagličić, S. Vrtnik, A. Bilušić and D. Stanić). Jovica's research has contributed importantly to over ten joint publications in respectable scientific journals (Physical Review B, Journal of alloys and compounds …)

Jovica Ivkov, Kristijan Velebit, Matej Bobnar (IJS, Ljubljana) and Ana Smontara (right to left) during a discussion of the results of measuring Hall effect of the *d*-AlNiCo quasicrystals (2010).

Jovica Ivkov was also a passionate reader of books and was especially interested in art, history and philosophy. His office library contained over a hundred books that he loved to read during long-term experiments. Jovica Ivkov had a good sense of design and he used it when he designed and set up the website of the *Laboratory for the physics of transport phenomena (http://condensed-matter.ifs.hr/)*, and designed websites and prepared posters and flyers for several scientific events (conferences and workshops) organized by the group of Ana Smontara.

Jovica Ivkov was a remarkably adept experimentalist for research of Hall effect, transferring with all his heart the knowledge to the younger scientists (contributions M. Čulo, P. Popčević and D. Starešinić). He was a valued collaborator and a great friend (contribution of E. Tutiš). His insight in metallic glasses, amorphous and complex metallic systems and-high temperature superconductors, his enthusiasm and curiosity will be missed by us, but he will be remembered by his many significant contributions.

Ana Smontara and Emil Babić

Program

10:00 – 10:15	Slobodan Milošević Ana Smontara	Welcome Address Obituary Jovica Ivkov
Chair: John R. Cooper		
10:15 – 10:30	Emil Babić	Hall effect and electronic structure of transition metal based metallic glasses
10:30 – 10:45	Ramir Ristić	Transport properties of $Cu_{55}Hf_{45-x}Ti_x$ metallic glasses
10:45 – 11:00	Mario Novak	Magnetoresistance in metallic glasses doped with hydrogen
11:00 – 11:15	Damir Starešinić	Low temperature heat capacity as the indicator of metallic glass properties
11:15 – 11:30	Coffee break	
Chair: Ivo Batistić		
11:30 – 11:45	Ante Bilušić	Thermoelectric properties of the *i*-Al-Cu-Fe quasicrystals
11:45 – 12:00	Denis Stanić	Hall effect measurements by the five-point compensation method of complex metallic alloys $Al_3(Mn,Fe)$
12:00 – 12:15	Janez Dolinšek	Hall effect in quasicrystals and complex intermetallics
12:15 – 12:30	Stane Vrtnik	Physical properties of the $Al_{13}Fe_4$ and related $Al_{13}(Fe,Ni)_4$ complex intermetallics
12:30 – 12:45	Zvonko Jagličić	Anisotropic physical properties of the Taylor-phase T-Al-Mn-Fe complex intermetallic
Chair: Pavo Dupček		
12:45 – 13:00	Nikola Radić	Phase transformations of aluminum-based thin films upon thermal annealing
13:00 – 13:15	Marija Sorić	Investigation of thermal stability of Al-Mo thin films
13:15 – 13:30	Ognjen Milat	Investigation of structural and physical properties of tungsten–carbon thin films prepared by reactive sputtering
13:30 – 13:45	Matija Čulo	Magnetotransport properties of $La_{1-x}Ca_xMnO_3$ ($0.52 \leq x \leq 0.75$): signature of phase coexistence
13:45 – 14:00	Petar Popčević	Magneto-transport properties of single crystal SnSe thermoelectric
14:00 – 14.15	Eduard Tutiš	Behind the Gateways of Anomalous Hall Effect

Hall effect and electronic structure of transition metal based metallic glasses

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The intimate relationship of the Hall coefficient, R_H, with the electronic band structure (EBS) and the dynamical properties of electrons in solids is particularly important in amorphous alloys, where most of the direct methods for the determination of the EBS are not applicable. Because of this, the results of the lifelong study of R_H in amorphous alloys (including thin films) by Dr. J. Ivkov, are one of the most important, permanent achievements from the over 40-year long extensive investigations of these systems in Zagreb. I will try to illustrate this by showing the correlations between R_H and the EBS in two most important types of metallic glasses (MG): late transition metal-metalloid (TL-M) and the early transition metal-late transition metal (TE-TE) MGs. In the former case (Ni-base MGs) Ivkov obtained accurate numbers of the conduction electrons per atom, n_a , over a broad composition range which enabled him to deduce changes in the EBS with the content and type of M[1]. He could also show that the Ziman-Faber theory can explain the transport properties of these MGs. Similarly, Ivkov was the first to demonstrate the correlation between the split-band EBS and the change of sign of R_H (Fig. 1) in TE-Tl MGs[2] . He also provided some evidence for the connection between positive values of R_H and the spin-orbit interaction in these alloys. Equally important was his study of anomalous Hall effect and the EBS in ferromagnetic TL-M MGs [3].

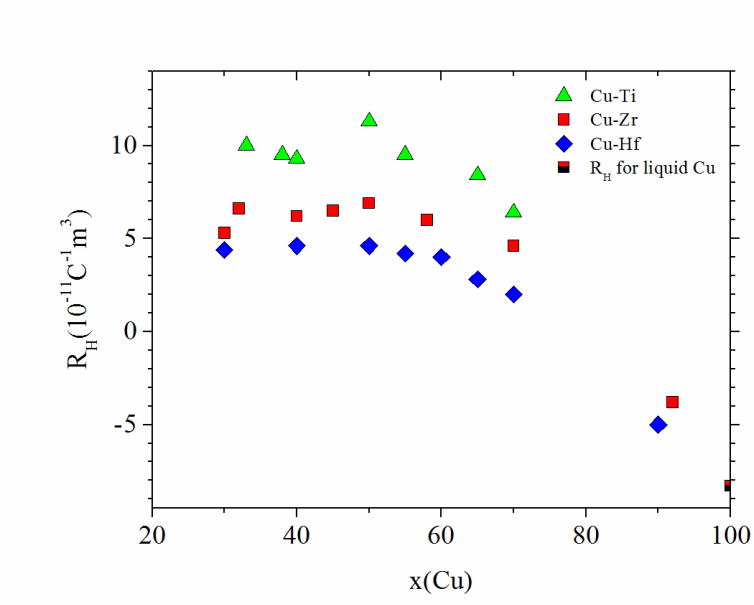

Fig.1. Hall coefficients of Cu-Ti,Zr,Hf alloys.

[1] **J. Ivkov**, E. Babic, J. Phys. F: Met. Phys. **15**, L161 (1985); J. Ivkov , E. Babic, H. H. Liebermann, J. Phys.:Cond. Mat. **2**, 8591 (1990)

[2] **J. Ivkov**, E. Babic, R. L. Jacobs, J. Phys. F: Met. Phys. **14**, L53 (1984); J. Ivkov, E. Babic, J. Phys.: Cond. Mat.**2**. 3891 (1990)

[3] **J. Ivkov**, Ž. Marohnić, P. Dubček, E. Babić, J. Phys. F: Met. Phys. **14**, 3023 (1984)

Transport properties of $Cu_{55}Hf_{45-x}Ti_x$ metallic glasses

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Recent studies of $Cu_{55}Hf_{45-x}Ti_x$ metallic glass ($0 \leq x \leq 45$) [1] indicate the formation of bulk metallic glass (BMG) for $15 \leq x \leq 30$. In the same compositional range ratio of the glass transition temperature T_g and the temperature of liquid state T_l shows slight maximum consistent with the greatest glass forming ability (GFA). A recent study of the magnetic susceptibility and mechanical properties (Young's modulus and microhardness) showed a uniform change of these properties with x without any special features in the compositional range where the BMGs form [2].

We will present the results of measurement of the electrical resistivity and the Hall coefficient of these alloys as a function of concentration and temperature ($80K \leq T \leq 300K$) [3]. These properties, closely related to the electronic structure and dynamics of electrons, monotonously depend on x and show no special features in the area of BMGs. The temperature dependence of resistances is consistent with the effects of the weak initial localization of electrons. The Hall coefficients (R_H) are positive, practically independent of temperature and grow approximately linearly with x. So, neither R_H nor electrical resistance indicates the formation of BMG. As studied alloys have the same number of electrons per atom e/a, it cannot be associated with BMG formation. [4]

[1] I. A. Figueroa, H. A. Davies, I. Todd, I. A. Verduzco, P. Hawksworth, Adv. In Tech. of Met. and Mat. Proc. J. **8** (2006) 146-151)

[2] R. Ristić, E. Babić, D. Pajić, K. Zadro, I. A. Figueroa, H. A. Davies, I. Todd, A. Kuršumović, J. Alloys Compd. **504S** (2010) S194-S197;

[3] **J. Ivkov**, R. Ristić, E. Babić and I. Figueroa, Transport properties of $Cu_{55}Hf_{45-x}Ti_x$ metallic glasses, to be published.

[4] M. Fukuhara, M. Takahashi, Y. Kawazoe, I. Inoue, Appl. Phys. Lett. **90** (2007) 073114.

Magnetoresistance in metallic glasses doped with hydrogen

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Magnetoresistivity of $(Zr_2(3d))_{1-x}H_x$ metallic glasses (where 3d stands for Fe and Co atoms) was investigated as a function of hydrogen doping (x) in the temperature range from 100 K down to 5 K [1]. Obtained magnetoresistivity is always positive due to the strong spin–orbit interaction and well described in the terms of exchange-enhanced spin-splitting contributions to the usual weak-localization term. $(Zr_2(Fe))_{1-x}H_x$ exhibits a significantly stronger magnetoresistivity than $(Zr_2(Co))_{1-x}H_x$ for all measured temperatures and doping levels, which is attributed to the increase in the inelastic spin-scattering rate (τ_{in}^{-1}) and the Stoner factor $(1-I)^{-1}$. The magnetoresistivity of $(Zr_2(3d))_{1-x}H_x$ exhibits a simple B^2 behavior at higher temperatures, providing the information on τ_{in}^{-1}, and a more complex behavior at low temperatures, which gives the information on the spin–orbit scattering rate τ_{so}^{-1}. It was found that the increase in the doping level reduces τ_{so}^{-1}, indicating hybridization of hydrogen *s*-electrons with Zr *d*-electrons and thus reducing the spin–orbit interaction

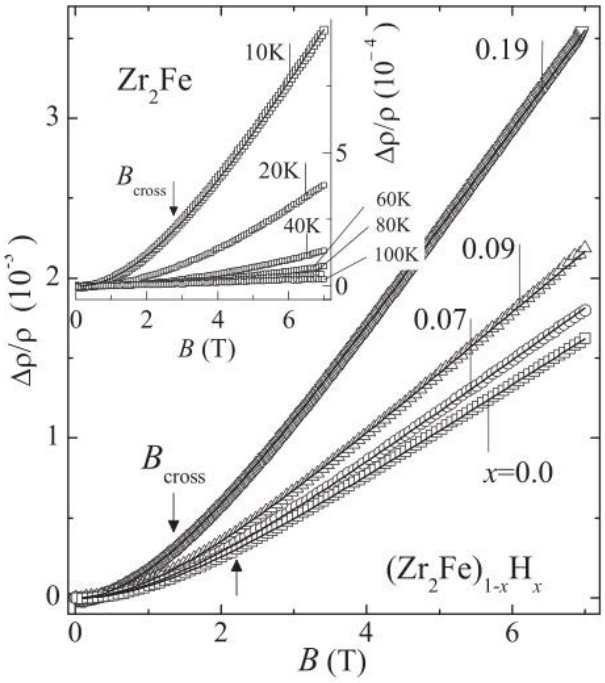

Fig. 1. (Main panel) The MR of $(Zr_2(Fe))_{1-x}H_x$ at T = 5 K for x = 0.0, 0.07, 0.09 and 0.19. The MR shows B^2 dependence for small fields and fairly linear dependence for larger fields (B > 2 T) and increases by increasing x. The vertical arrow marks B_{cross} , a maximal B for which the MR is still quadratic. (Inset) The MR of Zr_2Fe versus B at several different temperatures as indicated.

[1] M. Novak and I. Kokanović, Journal of Non-Crystalline Solids **376**, 86 (2013).

Low temperature heat capacity as the indicator of metallic glass properties

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We present the results of the low temperature heat capacity c_p measurements for several metallic glasses (MG) ranging from simple binary MG $Zr_{77}Ni_{23}$ [1], over bulk MG $Cu_{55}Hf_{45-x}Ti_x$ [2,3] to high entropy MG $(TiZrNbCu)_{1-x}Ni_x$ [4,5]. The electronic contribution γ is used to evaluate the density of states at the Fermi level N_F, while the acoustic contribution β gives the Debye temperature θ_D related to the elastic properties. The boson peak, although its origin is still a matter of debate, can be related to the structural heterogeneity of MG. A careful analysis is required to reliably determine these contributions.

The boson peak, which is considered universal feature of glasses, is absent in the low temperature c_p of $Zr_{77}Ni_{23}$ MG [1]. We argue that the boson peak depends on the sample history and it can disappear close to minimum cooling rates for glass formation. In $Cu_{55}Hf_{45-x}Ti_x$ neither N_F nor θ_D can be used as the indicator of glass forming ability (GFA) [2]. However, the boson peak correlates well with GFA [3], as shown in Figure 1., suggesting that it can be used as indicator of GFA for MG formed under comparable conditions.

The electronic and elastic properties of high entropy MG $(TiZrNbCu)_{1-x}Ni_x$ derived from low temperature c_p are consistent with other measurements and indicate very similar behavior as in binary and ternary early-late transition metal MG [4]. We show for the first time the boson peak in high entropy MG [4,5] which increases with Ni content, as shown in Figure 2.

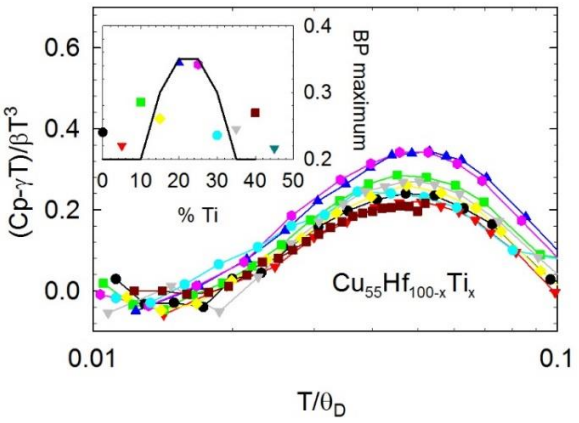

Figure 1. Boson peak in c_p/T^3 of $Cu_{55}Hf_{45-x}Ti_x$. In the inset is the boson peak maximum vs. maximum thickness of amorphous phase as a measure of GFA.

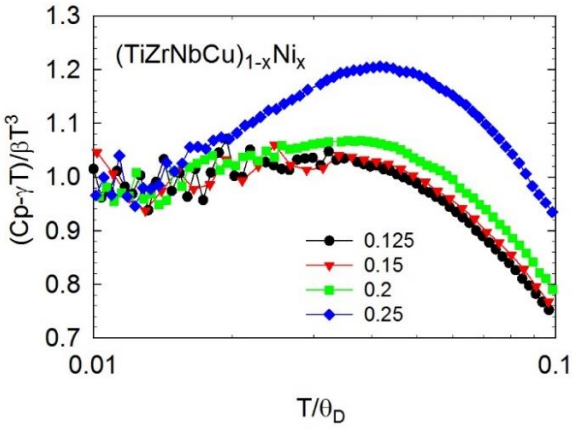

Figure 2. Boson peak in c_p/T^3 of $(TiZrNbCu)_{1-x}Ni_x$.

[1] A. Salčinović Fetić, S. Sulejmanović, G. Remenyi, D. Starešinić, A. Kuršumović, E. Babić, K. Biljaković, *Fragility of $Zr_{77}Ni_{23}$ metallic glass: evidence in heat capacity*, under consideration in Phys. Rev. B

[2] G. Remenyi, K. Biljaković, D. Starešinić, D. Dominko, R. Ristić, E. Babić, I. A. Figueroa, H. A. Davies, *Looking for footprint of bulk metallic glass in electronic and phonon heat capacities of $Cu_{55}Hf_{45-x}Ti_x$ alloys*, Appl. Phys. Lett. **104** (2014) 171906-1-4

[3] A. Salčinović Fetić, G. Remenyi, I. A. Figueroa, D. Starešinić, A. Kuršumović, R. Ristić, E. Babić, K. Biljaković, *Boson peak and glass forming ability of $Cu_{55}Hf_{45-x}Ti_x$ metallic glass*, under preparation

[4] K. Biljaković, G. Remenyi, I. A. Figueroa, R. Ristić, D. Pajić, A. Kuršumović, D. Starešinić, K. Zadro, E. Babić, *Electronic structure and properties of $(TiZrNbCu)_{1-x}Ni_x$ high entropy amorphous alloys*, J. Alloys Compd. **695** (2017) 2661-2668

[5] *The boson peak in $(TiZrNbCu)_{1-x}Ni_x$ high entropy amorphous alloys*, work in progress

Thermoelectric properties of the *i*-Al-Cu-Fe quasicrystals

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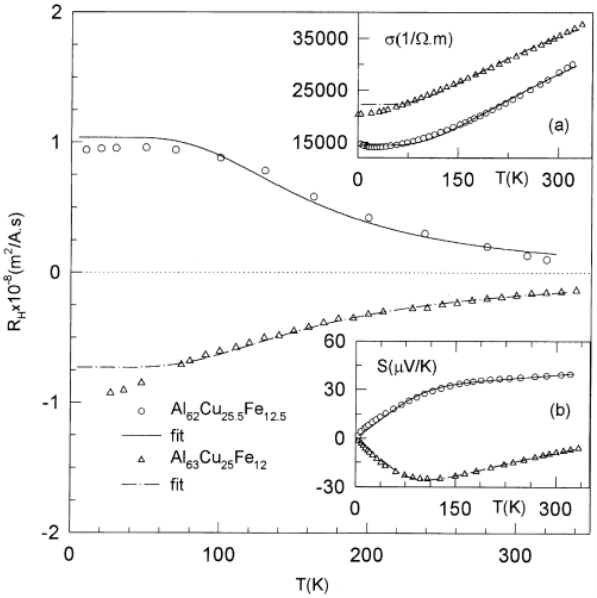
The beginning of the quasicrystals' research at the Institute of Physics, Zagreb coincided with the start of my work at the Institute. The very first quasicrystal system I met was icosahedral Al-Cu-Fe, which was topic of my Master thesis [1]. At that time, almost twenty years ago, sample production methods were not optimized enough to provide pure quasicrystal systems. However, our very promising results on i-Al-Cu-Fe enabled us in following almost two decades strong and successful networking with quasicrystal and complex metal alloys' community. Very important part of quasicrystal research at the Institute of Physics, starting from its' very beginning was Hall-effect laboratory led by our dear colleague Jovica Ivkov. Our collaboration, continued after I left the Institute too, resulted in several joint publications from the field of quasicrystals and complex metallic alloys [2-4].

In memory of Jovica, I will present experimental electric, thermoelectric and Hall-effect results of icosahedral Al-Cu-Fe quasicrystal system. In a temperature range between 4 K and 300 K we examined two members of the system: *i*-$Al_{62}Cu_{25.5}Fe_{12.5}$ and *i*-$Al_{63}Cu_{25}Fe_{12}$. Although of very similar composition, their transport properties differ considerably. The most noticeable are opposite signs of their thermoelectric power and Hall-coefficient, indicating different carrier types in these systems. The reason is composition balance between size of the pseudo-Brillouin zone and the Fermi sphere, as in Hume-Rothery alloys: a small change in composition varies the size of the Fermi sphere, while the pseudo Brillouin zone remains practically intact. Although due to existing pseudo-gap these systems are basically semi-metallic, for detailed numerical analysis we used two-band model known from theory of semiconductors (see figure, taken from Ref. [2]). Discrepancy of the fit and low-temperature electrical conductivity from data is explained by quantum interference effects.



[1] A. Bilušić, *Investigations of transport properties of Al-Cu-Fe quasicrystals* M. Sc. Thesis, University of Zagreb (1999).

[2] A. Bilušić; A.Smontara; J.C. Lasjaunias; J.**Ivkov**; Y.Calvayrac, *Thermal and thermoelectric properties of icosahedral $Al_{62}Cu_{25.5}Fe_{12.5}$ quasicrystal.* Materials science and engineering A, **294/296** (2000) 711-714.

[3] D. Stanić; P.Popčević; I.Smiljanić; A.Bilušić; I. Batistić; **J.Ivkov**; A. Smontara, *Thermal conductivity of Taylor phases T-$Al_{73}Mn_{27}$*, Journal of physics. Conference series. **226** (2010); 012034-1-012034-5.

[4] P. Popčević; K.Velebit; D. Stanić; **J. Ivkov**; Ž. Bihar; A.Bilušić; A.Smontara, *Thermoelctric properties of quasicrystals*, Book of abstracts and programme of the 14th Joint Vacuum Conference - 12th European Vacuum Conference - 11th Annual Meeting of the German Vacuum Society - 19th Croatian - Slovenian Vacuum Meeting 2012, Eds.: N. Radić, S.Milosevic and B. Pivac, 2012, Dubrovnik, Croatia.

Hall effect measurements by the five-point compensation method of complex metallic alloys $Al_3(Mn,Fe)$

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I started my PhD research ten years ago at the Laboratory for the Study of Transport Problems at the Institute of Physics, Zagreb. During my research, I was studying transport properties of complex metallic alloys. One of the important physical parameters in characterizing the electronic transport properties of metallic systems is the Hall coefficient. Dr. Jovica Ivkov introduced me to the five-point compensation method, for which I am very grateful to him. I have assisted him in the experimental determination of the normal and the anomalous Hall coefficients of many complex metallic alloys, quasicrystals and their approximants. Our collaboration has continued after I finished my thesis and has resulted in several joint publications.

In memory of Jovica I will present the Hall coefficient (R_H) determination of complex metallic alloys $Al_{73}Mn_{27-x}Fe_x$ (x = 0, 2, 4 and 6) [1]. All the alloys were *T* (Taylor) phase except $Al_{73}Mn_{21}Fe_6$ that was a decagonal (*d*) quasicrystal. The Hall coefficients of all the samples were positive and they decreased strongly with the increase of temperature according to the Curie-Weiss [C/(T–q)] law. Therefore, for the separation of the normal (R_0) and anomalous (R_S) Hall coefficients, the results for the paramagnetic susceptibility (χ) in the corresponding temperature interval have been used. The values deduced from the $R_H(\chi)$ plots were about $-2\cdot10^{-10}$ m^3 C^{-1} for R_0, and about $5\cdot10^{-7}$ m^3 C^{-1} for R_S. When the possible dependence of R_S on temperature is taken into account, that is due to the temperature dependence of the electrical resistivity $\rho(T)$, the values were about zero for R_0, and about $3\cdot10^{-7}$ m^3 C^{-1} for R_S (295 K). No significant composition dependence of R_0 was detected.

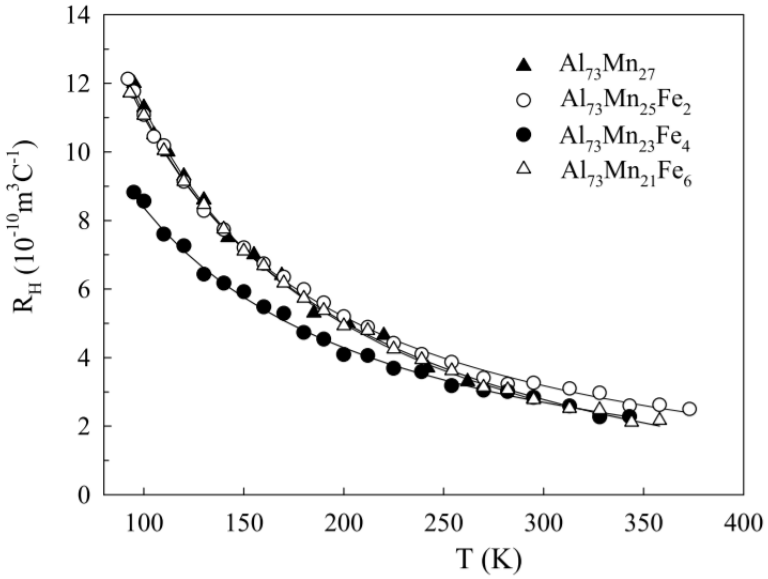

Fig. 1. Temperature-dependence of the Hall coefficient of $Al_{73}Mn_{27-x}Fe_x$.

[1] D. Stanić, **J. Ivkov**, A. Smontara, Z. Jagličić, J. Dolinšek, M. Heggen, M.Feuerbacher, *Hall effect in Taylor-phase and decagonal $Al_3(Mn,Fe)$ complex intermetallics*, Z. Kristallogr. **224** (2009) 49-52

Hall effect in quasicrystals and complex intermetallics

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The temperature- and magnetic-field dependent Hall coefficient is an important physical parameter to characterize electronic transport properties of metallic systems. During long-lasting scientific cooperation between the Solid State Physics Department of the Jožef Stefan Institute, Ljubljana, and the Laboratory for the Study of Transport Problems of the Institute of Physics, Zagreb, many intermetallic systems were jointly investigated, with the essential contribution of Dr. Jovica Ivkov. Among the investigated compounds, Dr. Ivkov has determined experimentally the normal and the anomalous Hall coefficients of the $Al_{80}Cr_{15}Fe_5$ and Y-Al-Ni-Co decagonal approximants, the $Al_{13}Co_4$, the T-Al_3(Mn,Fe), the $Al_{13}(Fe,Ni)_4$ and the δ-$FeZn_{10}$ complex metallic alloys, the *d*-Al-Ni-Co decagonal quasicrystal, the *i*-Au-Al-Yb icosahedral quasicrystal, and the GaPd and InPd intermetallic catalysts. The work of Dr. Ivkov has contributed importantly to eleven joint publications during 2007–2014, out of which seven were published in Phys. Rev. B. Dr. Ivkov was an extraordinarily skilled experimentalist for the field of Hall effect and he was gladly transferring the knowledge to younger generation, including PhD students and postroctoral fellows from the J. Stefan Institute. Though Dr. Ivkov has left us physically, the science he created will stay with us forever.

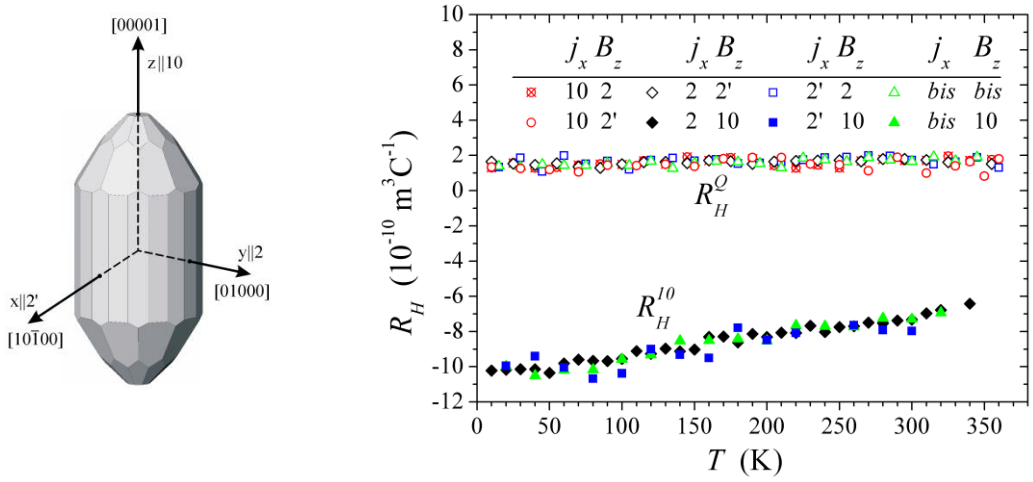

Figure 1: Temperature- and crystallographic-direction-dependent Hall coefficient of the decagonal *d*-Al-Ni-Co, determined by Dr. Jovica Ivkov (published in: *Intrinsic anisotropic magnetic, electrical and thermal transport properties of d-Al-Co-Ni decagonal quasicrystal*, M. Bobnar, P. Jeglič, M. Klanjšek, Z. Jagličić, M. Wencka, P. Popčević, **J. Ivkov**, D. Stanić, A. Smontara, P. Gille, J. Dolinšek, Phys. Rev. B **85**, 024205 (2012)).

Physical properties of the $Al_{13}Fe_4$ and related $Al_{13}(Fe,Ni)_4$ complex intermetallics

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We have investigated the magnetic susceptibility, the electrical resistivity, the specific heat, the thermoelectric power, the Hall coefficient, and the thermal conductivity of the $Al_{13}Fe_4$ and $Al_{13}(Fe,Ni)_4$ monoclinic approximants to the decagonal quasicrystal [1]. While the $Al_{13}Fe_4$ crystals are structurally well ordered, the ternary derivative $Al_{13}(Fe,Ni)_4$ contains substitutional disorder and is considered as a disordered version of the $Al_{13}Fe_4$. The crystallographic-direction-dependent measurements were performed along the a^*, b, and c directions of the monoclinic unit cell, where the (a^*, c) atomic planes are stacked along the b direction. The electronic transport and the magnetic properties exhibit significant anisotropy. The stacking b direction is the most conducting direction for the electricity and heat. The effect of substitutional disorder in $Al_{13}(Fe,Ni)_4$ is manifested in the large residual resistivity $\rho(T\rightarrow 0)$ and significantly reduced thermal conductivity of this compound, as compared to the ordered $Al_{13}Fe_4$. Specific-heat measurements reveal that the electronic density of states at the Fermi level of both compounds is high. The anisotropic Hall coefficient R_H reflects complex structure of the anisotropic Fermi surface that contains electronlike and holelike contributions. Depending on the combination of directions of the current and the magnetic field, electronlike ($R_H < 0$) or holelike ($R_H > 0$) contributions may dominate, or the two contributions compensate each other ($R_H \approx 0$). Similar complicated anisotropic behavior was observed also in the thermopower.

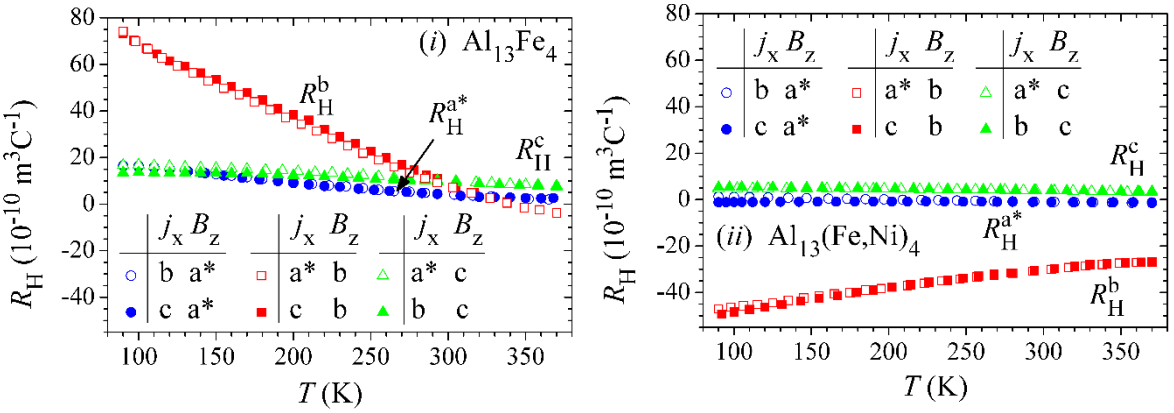

Figure 1. Anisotropic temperature-dependent Hall coefficient $R_H = E_y / j_x B_z$ for different combinations of directions of the current j_x and magnetic field B_z. The superscript a^*, b, or c on R_H denotes the direction of the magnetic field. The Hall coefficients of the $Al_{13}Fe_4$ and the $Al_{13}(Fe,Ni)_4$ show complicated temperature-dependent anisotropy for different combinations of the current and field directions.

[1] P. Popčević, A. Smontara, **J. Ivkov**, M. Wencka, M. Komelj, P. Jeglič, S. Vrtnik, M. Bobnar, Z. Jagličić, B. Bauer, P. Gille, H. Borrmann, U. Burkhardt, Y. Grin, J. Dolinšek, Phys. Rev. B **81**, 184203 (2010)

Anisotropic physical properties of the Taylor-phase T-Al-Mn-Fe complex intermetallic

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The orthorhombic Taylor phase T-$Al_{72.5}Mn_{21.5}Fe_{6.0}$ is built of two atomic layers within (*a*, *c*) planes stacked along the *b* pseudotenfold crystallographic axis, a flat layer, and a pucked layer composed of two sublayers. The unit cell contains 156 atoms and can be consider as an approximant to the decagonal Al-Mn-Pd quasicrystal. [1]

The magnetic properties of an oriented single crystal T-Al-Mn-Fe are similar to the polygrain sample and practically isotropic. The compound shows a spin-glass-like transition at 29 K (Fig. 1) and was employed to construct a new kind of memory element: a thermal memory cell.

The electrical resistivity is the lowest along the *b* direction. The thermopower and Hall-coefficient measurements (Fig. 2) demonstrate the holes are the dominant charge carriers. The anisotropy of the thermal conductivity is practically negligible. The T-Al-Mn-Fe Taylor phase can be considered as almost isotropic complex intermetallic.

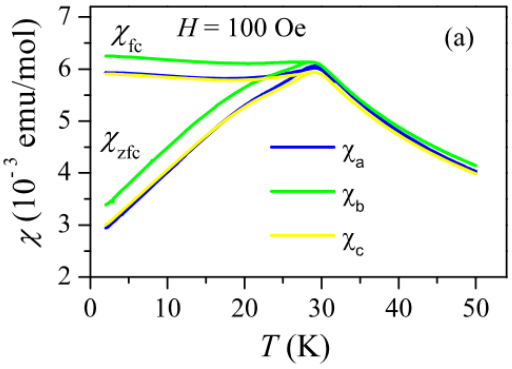

Fig. 1 Magnetic susceptibility with external magnetic field along *a*, *b* and *c* directions

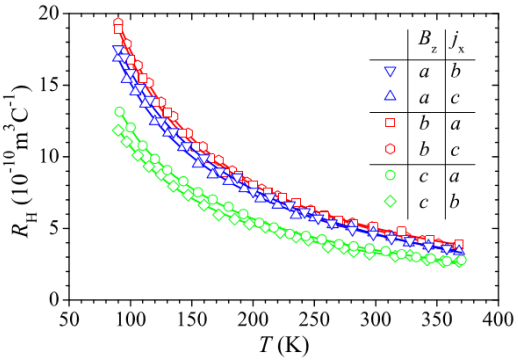

Fig. 2 Anisotropic temperature-dependent Hall coefficient

The electrical resistivity is the lowest along the *b* direction. The thermopower and Hall-coefficient measurements demonstrate the holes are the dominant charge carriers. The anisotropy of the thermal conductivity is practically negligible. The T-Al-Mn-Fe Taylor phase can be considered as almost isotropic complex intermetallic.

[1] M. Heggen, M. Feuerbacher, **J. Ivkov**, P. Popčević, I. Batistić, A. Smontara, M. Jagodič, Z. Jagličić, J. Janovec, M. Wencka, J. Dolinšek, Phys. Rev. B **81**, 184204 (2010) .

Phase transformations of aluminum-based thin films upon thermal annealing

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The amorphous alloys of aluminum and refractory transition metals, RM, are very interesting both from scientific point of view and for practical applications in modern technologies. These alloys exhibit very good mechanical and corrosion properties, and can be used as a diffusion barriers in integrated microelectronic circuits. Amorphous alloys can also serve as a precursor for production (by the heat treatment) of novel nanocrystalline or amorphous matrix-nanocrystalline materials. However, being in a thermodynamically metastable state, amorphous alloys tend to transform to more stable states depending on circumstances.

Normal thermal equilibrium solubility of transition metals in aluminium being rarely more than about 1 at.%, it is necessary to resort to non-equilibrium alloying methods to prepare these metastable alloys. Thin films of binary Al-RM (Mo, Nb, Ta, W, and V) alloys have been prepared by co-deposition from two independently controlled magnetron sources in a wide range of chemical composition. The as-prepared films were amorphous in a wide range of composition. Thermal stability of the prepared Al-RM films has been examined by isochronal (2 K/min) annealing in vacuum (10^{-4} Pa) up to 800 °C, and continuous monitoring of the electrical resistance of the film – Jovica Ivkov performed these extensive measurements with meticulous care and analyzed them in comprehensive depths [1-3]. Strong variation of electrical resistance with temperature (Fig.1) indicated onset of phase transformations in the film. The new phases were observed and identified by the XRD method at selected annealing temperatures. A summarized presentation of the crystallization temperatures (T_c) for the Al-RM amorphous alloys is shown in Fig.2.

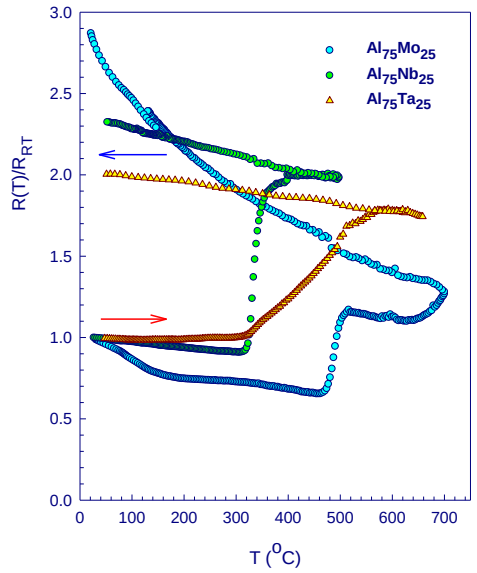

Fig.1

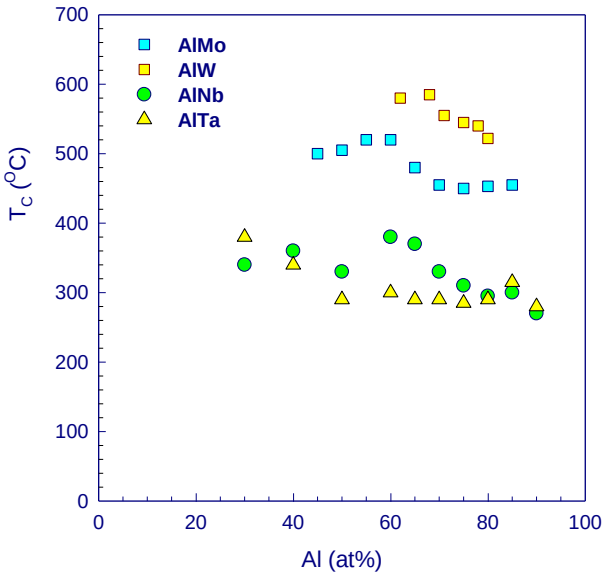

Fig.2

[1] **J. Ivkov**; K. Salamon; N. Radic; M. Soric: *Thermal stability of Al-Mo thin film alloys*, Journal of alloys and compounds **646** (2015) 1109-1115.

[2] **J. Ivkov**; N. Radic; A.Tonejc, A; T. Car, *Structural relaxation of Al-W amorphous thin films*, Journal of non-crystalline solids **319** (2003) 232-240.

[3] **J. Ivkov**; N. Radić, *The electrical resistivity in Al-W amorphous alloys*, Solid state communications **106** (1998) 273-277.

Investigation of thermal stability of Al-Mo thin films

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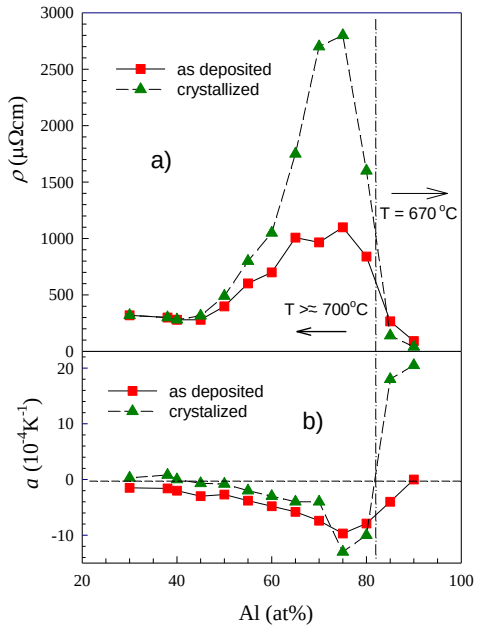
Investigation of thermal stability thin film is part of longstanding and successful collaboration between dr. Jovica Ivkov and dr. Nikola Radić (Laboratory for Thin Films, Institute Ruđer Bošković), whereas dr. Krešimir Salamon joined this collaboration performing structural characterization.[1] Preparing seminar research included in doctoral studies on the topic "Investigation of electrical resistivity of Al-Mo thin films", dr. Ivkov invited me to collaborate during the investigation of transport properties of thin films, so in memory of dr. Jovica Ivkov, I will present results of investigation of thermal stability of Al_xMo_{100-x} thin films.[2]

Thin Al_xMo_{100-x} films ($40 \leq x \leq 90$ with x in steps of 5 at % Al) were prepared by magnetron co-deposition onto alumina, glass, and sapphire substrates at room temperature. The film thickness was about 400 nm, and they were amorphous for $45 \leq x \leq 85$. The films' structural changes upon heating were investigated by measurement of the electrical resistivity variation with temperature, $\rho(T)$, during the isochronal heating. Thus obtained results were complemented, and conclusions confirmed, by GIXRD analysis for selected heating temperatures. The dynamical temperatures of crystallization, T_x, were determined from the sharp increase of the derivative of ρ with respect to temperature. No systematic dependence of T_x on film substrate has been observed. Except for the $Al_{85}Mo_{15}$ film, the ρ of the amorphous films increase on the crystallization. The temperature of crystallization exhibits maximum around 530°C for alloy compositions with x = 55 and 60. Electrical resistivity of both amorphous and crystallized films show a strong dependence on alloy composition, with a maximum for $Al_{75}Mo_{25}$ alloy. The resistivity of $Al_{75}Mo_{25}$ film is very large and amounts to 1000 μΩcm and 3000 μΩcm in amorphous and crystallized film, respectively, with the large negative temperature coefficient of -10×10^{-4} K^{-1} and -14×10^{-4} K^{-1}, respectively (Fig.).



Although the crystallization temperature observed for the examined amorphous Al-Mo alloys is not very high, it might allow to exploit excellent corrosion properties of such films at some elevated temperatures.

[1] **J. Ivkov**, N. Radić, K. Salamon, M. Sorić, *Thermal stability of Al-Mo thin film alloys*, Journal of alloys and compounds, **646** (2015) 1109-1115.

[2] **J. Ivkov**, N.Radić, K. Salamon; M. Sorić, *Investigation of thermal stability of Al-Mo thin films*, 16th International Conference on Thin Films Programme and book of abstracts, N. Radić and H. Zorc, (Eds.). Zagreb : Croatian Vacuum Society, 2014. p. 104.

Investigation of structural and physical properties of tungsten–carbon thin films prepared by reactive sputtering

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I collaborated with Jovica Ivkov on a number of research projects, and in particular on structural and physical properties of Tungsten–carbon thin films which had been deposited from argon–benzene discharges during DC magnetron, by Nikola Radić [1].

Deposition onto glass, monocrystalline silicon, tantalum and stainless steel at room temperature yielded W–C films, having XRD patterns corresponding to the structure of heavily disordered W_2C or β-WC_{1-x} carbides, Fig. 1. The samples deposited upon the Au or Cu foils were nanocrystalline cubic WC_{1-x} with the grain size of 2.9 nm. Disordered tungsten–carbon films were stable up to 1200°C. Microhardness of the films with disordered W_2C phase was about 5–6 GPa while that of the films with disordered β-WC_{1-x} phase was about 17 GPa.

The characteristics of films can be understood considering the effects of the incorporation of free carbon and/or carbon–hydrogen fragments into the tungsten carbide layer.

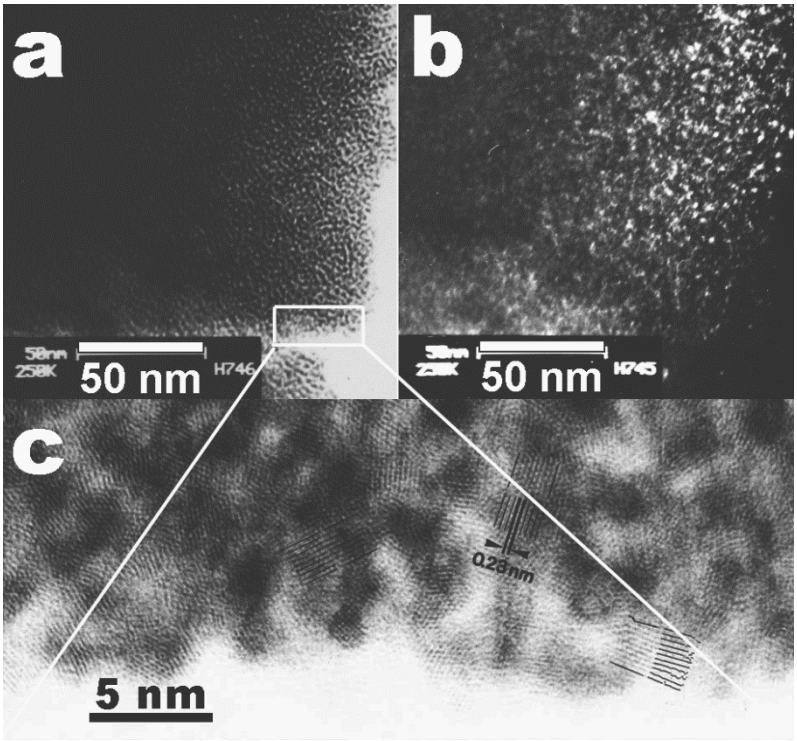

Fig 1. Imaging of W-C thin film consisting of dense aggregates of β-WC_{1-x} nanocrystals; (a) BF, (b) DF, (c) HR; position of objective aperture for (b) is indicated by circle in Fig 2(b). Particles are visible as dark dots in (a) or bright dots in (b), while their crystalline feature is revealed by 0.23 nm lattice fringes in (c). Patches consisting of 10-13 parallel fringes running in all directions are consistent with the average crystallite size of 2.8 nm, uniform spherical shape, and isotropic orientation.

[1] N. Radić, B. Gržeta, O. Milat, **J. Ivkov**, M. Stubičar, *Tungsten–carbon thin films prepared by reactive sputtering from argon–benzene discharges*, Thin Solid Films **320** (1998) 192–197.

Magnetotransport properties of $La_{1-x}Ca_xMnO_3$ ($0.52 \leq x \leq 0.75$): signature of phase coexistence

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I will present the temperature and magnetic field dependence of transport properties in epitaxial films of the manganite $La_{1-x}Ca_xMnO_3$ in the overdoped region $x > 0.5$ of the phase diagram shown in Figure 1, where a charge–ordered (CO) and an antiferromagnetic (AF) phase are present [1]. Resistivity, magnetoresistance and angular dependence of magnetoresistance were measured in the temperature interval 4.2 K < T < 300 K, for three concentrations x = 0.52, 0.58 and 0.75 and in magnetic fields up to 5 T. The semiconducting/insulating–like behavior in zero field was observed in the entire temperature range for all three concentrations x and the electric conduction in the CO state obeys 3D Mott's variable–range hopping model. A huge negative magnetoresistance for x = 0.52 and 0.58, a metal–insulator transition for B > 3 T for x = 0.52 and the presence of anisotropy in magnetoresistance for x = 0.52 and 0.58 show the fingerprints of colossal magnetoresistance (CMR) behavior implying the existence of ferromagnetic (FM) clusters. The declining influence of the FM clusters in the CO/AF part of the phase diagram with increasing x contributes to a possible explanation that a phase coexistence is the origin of the CMR phenomenon.

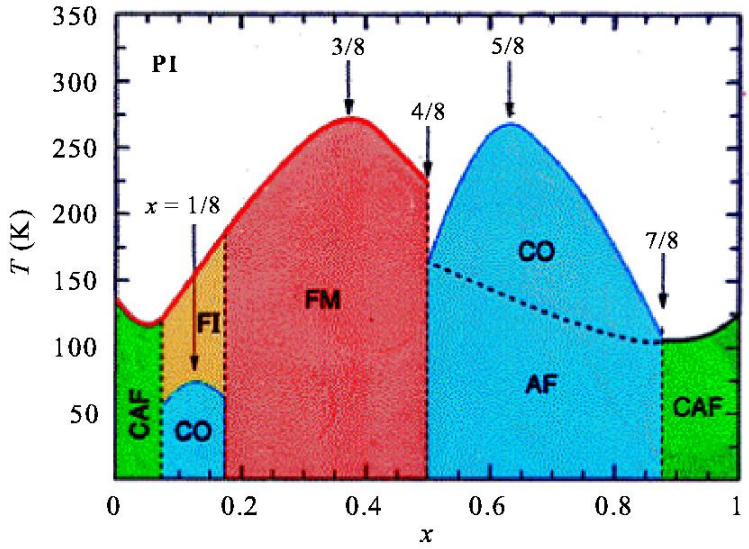

Fig. 1. Phase diagram of manganites $La_{1-x}Ca_xMnO_3$.

[1] M. Čulo, M. Basletić, E. Tafra, A. Hamzić, S. Tomić, F. Fischgrabe, V. Moshnyaga, B. Korin-Hamzić, *Magnetotransport properties of $La_{1-x}Ca_xMnO_3$ ($0.52 \leq x \leq 0.75$): signature of phase coexistence,* submitted to Thin Solid Films

Magneto-transport properties of single crystal SnSe thermoelectric

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Most of this work is inspired by knowledge acquired during long-term collaboration with Jovica Ivkov on research of Hall effect of quasiperiodic crystals and complex metallic alloys [1-4]. It was planned that Jovica takes part in investigation of magneto-transport properties of SnSe thermoelectric [5] as well, and I am missing his support.

Intermetallic compound SnSe is long known layered system whose enormous thermoelectric potential was revealed just recently [5]. In this system, thermoelectric figure of merit ZT reaches its peak value of 2.62 at 923 K. However, this high ZT value is realized only in high-temperature phase that exists above 750 K. In low temperature phase, SnSe has considerably smaller ZT value.

As a first step in the study of this system, we have experimentally examined magneto-transport in SnSe single crystals at temperatures below room temperature that is poorly covered in literature. At temperatures above 100 K electrical resistivity it exhibits T^2 like behavior. Seebeck coefficient shows linear behavior but with considerable residual term. At lower temperatures ($T < 70$ K) electrical resistivity exhibit semiconducting behavior but Seebeck coefficient although changing slope retains linear-like metallic behavior. At lowest temperatures where hoping like resistivity prevails, Hall resistivity shows nonlinear behavior and magnetoresistance changes sign from negative at low fields to positive at higher fields.

[1] A. Smontara, I. Smiljanić, **J. Ivkov**, D. Stanić, O.S. Barišić, Z Jagličić, P. Gille, M. Komelj, P. Jeglič, J. Dolinšek, *Anisotropic magnetic, electrical and thermal transport properties of Y-Al-Ni-Co decagonal approximant,* Physical Review B. **78** (2008) 104204.

[2] M. Komelj, **J. Ivkov**, A. Smontara, P. Gille, J. Dolinšek, *Origin of the Hall-coefficient anisotropy in the Y-Al-Ni-Co decagonal approximant,* Solid State Communications. **149** (2009) 515.

[3] J. Dolinšek, M. Komelj, P. Jeglič, S. Vrtnik, D. Stanić, P. Popčević, **J. Ivkov**, A. Smontara, Z. Jagličić, P. Gille, Y. Grin, *Anisotropic magnetic and transport properties of the orthorhombic Al13Co4 decagonal approximant,* Physical Review B. **79** (2009) 184201.

[4] S. Jazbec, S. Vrtnik, Z. Jagličić, S. Kashimoto, **J. Ivkov**, P. Popčević, A. Smontara, H.J. Kim, J.G. Kim, J. Dolinšek, *Electronic density of states and metastability of icosahedral Au-Al-Yb quasicrystal,* Journal of alloys and compounds. **586** (2014) 343.

[5] P. Popčević, M. Sorić, Peter Gille, N. Barišić, A. Smontara, *Magneto-transport properties of single crystal SnSe thermoelectric*, C-MAC Days 2016, Program and Abstracts,. Eds: Marc de Boissieu, Marek Mihalkovič Peter Švec, VEDA, Publishing House of the Slovak Academy of Sciences, Bratislava, Slovakia, November 21 -24, 2016, p. C-14. and references therein.

Behind the Gateways of Anomalous Hall Effect

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To the memory of our colleague Dr. Jovica Ivkov

The professional interests of Jovica and myself have overlapped in several instances, and I would like to recall some recent ones. Moreover, being colleagues at the Institute for long years, I had the opportunity to interact frequently with Jovica on "ordinary" level. I profited a lot from these contacts, as they went beyond ordinary in almost every occasion, basically because Dr. Jovica Ivkov was a person of considerable depth and richness that went in many directions. This depth and richness is not something that he was wearing and exposing without being asked. For newcomers, it required some attention to grasp and profit from what he cared for. I would like to recall few recent examples which show how his work and spirit affected my days and how they continue to do so for many of us.

Eduard Tutiš, Petar Popčević, **Jovica Ivkov** and Neven Barišić (right to left) discussing at the start of the UKF project 1B#65/10 *New electronic states driven by frustration in layered materials* (2010)