

Inductive-Type properties of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{100-x}$ Nanocomposites Produced by the Ion-Beam Sputtering in the Argon and Oxygen Ambient

T.N. Koltunowicz^{1,*}, P. Zhukowski^{1,†}, J.A. Fedotova^{2,‡}, A.V. Larkin^{3,§}

¹ Lublin University of Technology, 38a, Nadbystrzycka Str., 20-618 Lublin, Poland

² National Center for Particles and High Energy Physics of Belarusian State University, 153, Bogdanovich Str., 220040 Minsk, Belarus

³ Belarusian State University, 4, Independence Av., 220030 Minsk, Belarus

(Received 26 September 2011; published online 14 March 2012)

The paper discusses changes in the phase-shift angle Θ and capacity C_p depending on the frequency of alternating current of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposites characterized by the metallic phase content of $x = 38,2$ at.%. The tested samples have been produced in the argon and oxygen atmosphere by means of ion-beam sputtering of two targets made of the CoFeZr alloy and strips of the Al_2O_3 dielectric. Samples have been tested before and after their annealing at the temperature T and frequency f . Dependences of the phase-shift angle $\Theta(T, f)$ and capacity $C_p(T, f)$ have been determined for temperatures of the 77-373 K range and frequency values ranging from 50 to 10^6 Hz.

For the tested samples of the $x = 38,2$ at.% content it has been found that at high annealing temperatures the phase-shift angle is $\Theta < 0$ within the low-frequency area, while at high frequency values $\Theta > 0$. The analysis has shown that in the samples studied the series connection of capacity and noncoil-like inductance can be realized. Capacitive properties of LC circuit with $-90 \leq \Theta_L < 0^\circ$ are presented at low frequencies and its inductive properties with $0^\circ \leq \Theta_H < 90^\circ$ become apparent at high frequencies.

Keywords: Nanoparticles, Ion Implantation, Annealing, Measurement of Electrical Properties.

PACS numbers: 73.22. - f, 68.55.Ln, 81.40.Ef, 84.37.+q

1. INTRODUCTION

Materials of the authors' interest are granular metal-dielectric compounds of the metallic phase content x ranging from 30 at.% to 65 at.%, that are nanocomposites, which means that they include metallic-phase $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})$ spherical particles of the 6-10 nm dimensions randomly distributed in a dielectric (Al_2O_3) matrix [1, 2]. The two-phase metal-dielectric nanocomposites have been produced by ion-beam sputtering of two identical targets each one made of a CoFeZr plate with dielectric (Al_2O_3) strips fixed to it [1, 3]. Such configuration of a target makes possible to produce within one technological process thin layers (films) of the 1-6 nm thickness and of varied metallic phase-to-dielectric ratio. A mixture of argon of the $5,19 \cdot 10^{-2}$ Pa pressure and oxygen of the $9,6 \cdot 10^{-2}$ Pa pressure has been applied to the deposition process.

Earlier research works concerning nanocomposites of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ have presented a number of testing series performed with the application of alternating current and aiming at the determination of electric and magnetic properties of the discussed materials. Investigations concerning determination of temperature dependences of conductivity have been presented in [4, 5], a model of hopping conductivity – in [6, 7] and magnetic measurements performed with the application of electron paramagnetic resonance (EPR) – in [8].

The present article discusses results of testing $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposites of the metallic-phase content of $x = 38,2$ at.% that has been per-

formed at a measuring stand [9, 10]. The measurements have been performed with the application of alternating current of the frequency ranging from 50 Hz to 1 MHz and within the temperature interval from 77 K to 373 K. Each measurement series has been followed by 15 minute isochronous annealing of the samples in the temperature up to 873 K with the step of 25 K.

The main objective of the presented research project has been to determine frequency and temperature dependences of the phase-shift angle Θ and capacity C_p in granular nanocomposites and to present methods for producing noncoil-like inductance and a series capacity-inductance circuit.

2. THE PHENOMENON OF POSITIVE PHASE-SHIFT ANGLE OCCURRENCE IN $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ NANOMATERIALS

It has been observed that in the tested nanomaterials of the metallic-phase content x of not less than 38,2 at.%, produced by sputtering with a combined beam of $\text{Ar}^+\text{-O}^-$ ions and annealed in temperatures higher than 373 K over the 15-minute period, within the low-frequency area the phase-shift angle assumes negative values of $-90^\circ < \Theta < 0^\circ$, while in the area of high frequency values its value gets positive – $0^\circ < \Theta < +90^\circ$.

An analysis of $\Theta(f)$ characteristics presented in Fig. 1 has shown that similar phase-shift angle vs. frequency dependences occur in conventional circuits that

* t.koltunowicz@pollub.pl

† pawel@elektron.pol.lublin.pl

‡ julia@hep.by

§ LarkinAV@bsu.by

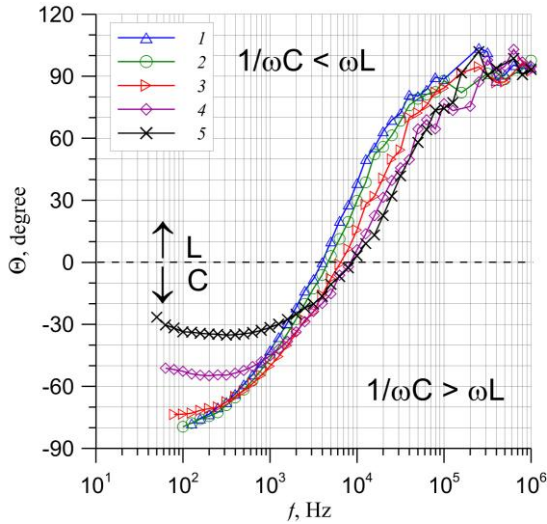


Fig. 1 – Phase-shift angle Θ vs. frequency for a $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ sample of $x = 38,2$ at.%, annealed at the temperature of $T_a = 823$ K. Measuring temperatures T_p : 1 – 153 K; 2 – 178 K; 3 – 218 K; 4 – 258 K; 5 – 293 K

include in-series connected discrete *RLC* elements i.e. that for such circuits in low-frequency areas $-90^\circ < \Theta_L < 0^\circ$, in high-frequency areas $0^\circ < \Theta_H < +90^\circ$, while $\Theta_R = 0^\circ$ at the resonance frequency described by the formula

$$f_R = \frac{1}{2\pi\sqrt{LC}} \quad (1)$$

It seems that similarity of $\Theta(f)$ characteristics recorded during the testing of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ materials produced by sputtering with the beam of Ar^+O_2^- ions and the ones obtained for a series *RLC* circuit is rather of a qualitative character as cases that have been investigated within the discussed project indicate that there are many features that are different for a nanocomposite film and for a series *RLC* circuit.

The first and the most important difference is that a nanocomposite layer does not include any winding, which is characteristic for the traditional inductance *L*.

Secondly, as can be seen in Fig. 1, in the low-frequency area a distinct $\Theta(f)$, minimum occurs, which cannot be found in the case of a *RLC* circuit.

Thirdly, resonance frequency f_R varies along with the measuring temperature T_p changes (Fig. 1), which means that one or both “elements” of a conventional circuit, *L* and *C* (1), change their values along with temperature changes. As f_R increases together with the growing temperature then, according to the formula (1), inductance or capacity or both of them should decrease along with the increase of the measuring temperature T_p .

Fig. 1 presents frequency dependence of capacity C_p for a sample of $x = 38,2$ at.%, annealed at the temperature of $T_a = 823$ K, whose phase-shift angle has been presented in Fig. 1.

As can be seen in Fig. 2, in the low-frequency area $C_p(f)$ rapidly decreases and reaches its minimal value at the frequency f_{\min} that is a function of the measuring temperature T_p . At further frequency growth a weak capacity increase can be observed.

As it has been established in [11], in the case of hopping recharging between two neutral potential wells ad-

ditional thermally activated dielectric permittivity occurs. An electron jumps from one neutral well to the other, which results in the formation of a dipole. After the τ time lapse the electron returns to the initial well and the dipole vanishes away. The frequency range, where the additional polarization occurs, is determined by the time τ and is included within the interval up to $1/2\tau$.

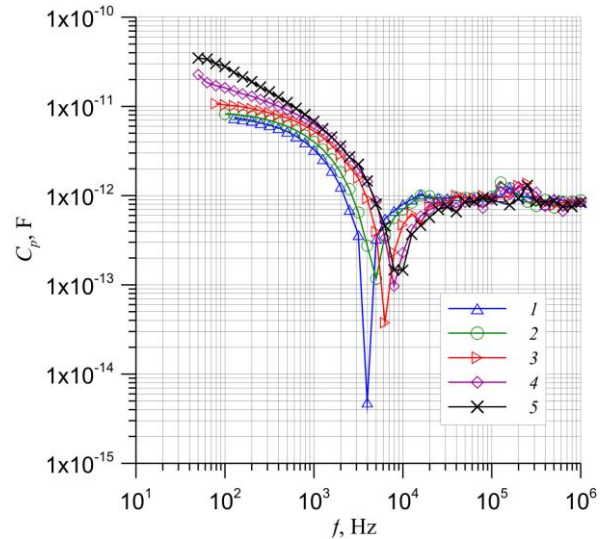


Fig. 2 – Frequency dependence of capacity C_p for a $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ sample of $x = 38,2$ at.%, annealed at the temperature $T_a = 823$ K. Measuring temperatures T_p : 1 – 133 K; 2 – 153 K; 3 – 178 K; 4 – 218 K; 5 – 258 K; 6 – 293 K

Fig. 3 presents a comparison of frequency dependences of the phase-shift angle Θ and capacity C_p for a sample of $x = 38,2$ at.%, annealed at the temperature of 823 K and tested at the temperature $T_p = 178$ K. It follows from the comparison that the position of f_{\min} precisely corresponds to the resonance frequency f_R , where the Θ angle value crosses the zero point (see: Fig. 1).

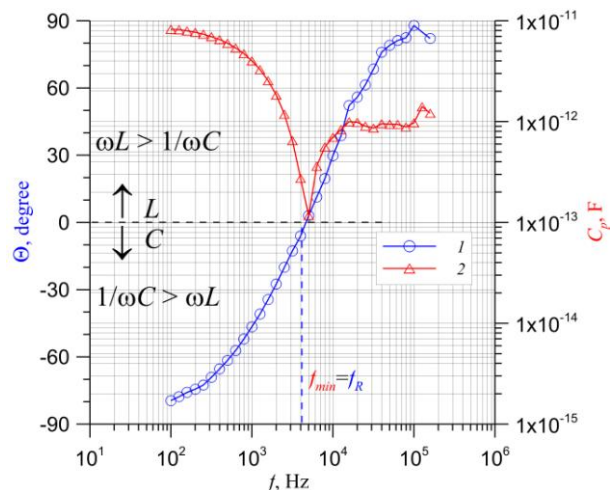


Fig. 3 – Comparison of frequency dependences of the phase-shift angle Θ (1) and capacity C_p (2) for a $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ sample of $x = 38,2$ at.%, annealed at the temperature of $T_a = 823$ K and measured at the temperature of $T_p = 178$ K

It has been shown in [12] that the resonance frequency f_R and the minimal frequency value f_{\min} overlap over the whole range of the applied measuring temperatures

T_p . The determined [see: 12] low values of activation energy f_{\min} indicate that the formation of noncoil-like inductance is related to the return jumps

of electrons from the potential well of a negative charge to the one of a positive charge that are determined by the time τ and characterized by low activation energy values.

For the tested nanocomposite samples, it has been observed that positive values of the phase-shift angle, that also occur in conventional RLC circuits, are related to the mechanism of hopping recharging. An electron jump from one neutral potential well to the other is realized in a time of the 10^{-13} s order and the return jump – after a lapse of time τ of the 10^{-3} - 10^{-6} s order. At the frequency values higher than $1/2\tau$ the phase lag $\omega\tau$ can reach more than 2π , which is favorable for the occurrence of positive phase-shift angles.

3. METHODS FOR PRODUCING NONCOIL-LIKE INDUCTANCE L AND A SERIES LC CIRCUIT BASED ON $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ NANOMATERIALS

The performed testing of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposites produced by $\text{Ar}^+\text{-O}_2^-$ ion-beam sputtering of targets composed of the $\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10}$ alloy and strips of the Al_2O_3 dielectric has given an important observation that in those materials positive phase-shift angles occur, which involves the formation of noncoil-like inductance. Based on the testing results two patent applications concerning the production of noncoil-like inductance areas [13] and series capacity-inductance circuits [14] to be applied in microelectronic systems have been filed.

The only hitherto known solutions are presented in the book [15] and the patent [16]. These are thin-layer inductive components with flat winding of the spiral or rectangular-meander shape and a magnetic core made of ferromagnetic film located at a parallel plane to the coil plane. Only that type of coils of a dielectric or magnetic base in the form a plane-uniform layer have been applied.

The above solutions yield low values of inductance per surface unit area and the inductive component occupies much space within a microelectronic circuit, which decreases its integration rate and brings about the occurrence of a considerable leakage flux that is characteristic for flat spiral winding.

Fig. 4 presents a method for producing noncoil-like inductance areas and a series capacity-inductance circuit in the $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposite. It is included in the [13] patent application that ion-beam sputter deposition of a ferromagnetic film composed of $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_{38,2}(\text{Al}_2\text{O}_3)_{61,8}$ in the argon-oxygen atmosphere at the pressure of $5,19 \cdot 10^{-2}$ Pa and $4,41 \cdot 10^{-2}$ Pa, respectively, onto a silicon substrate that has been earlier subdued to all technological procedures required at the microelectronic circuit production and then to 15-minute stabilizing annealing in the temperature of 848 K makes possible to produce noncoil-like inductance areas for microelectronic circuits. Induction of the obtained noncoil-like inductance inductance is of the $20 \mu\text{H}/\mu\text{m}^3$ order. The resulting advantage is that the

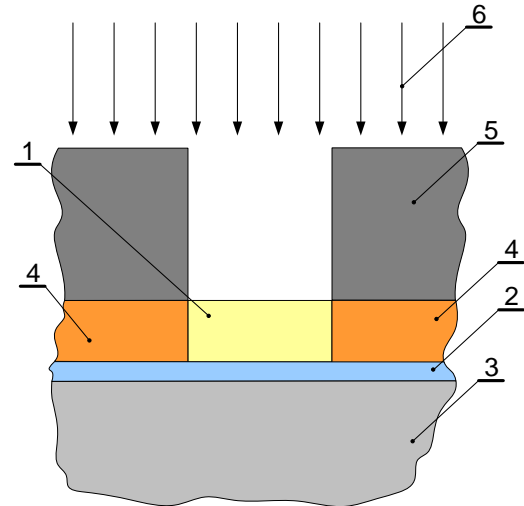


Fig. 4 – A method for producing noncoil-like inductance areas and a series capacity-inductance circuit in the $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposite. 1 – vapor-deposited film of the $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposite, 2 – insulation layer 3 – base silicon plate, 4 – metallic contacts, 5 – photomask, 6 – flux of sputtered atoms

semiconductor structure surface can be reduced and its integration rate can be increased.

The [14] patent application includes the same type of procedures only that the 15-minute annealing is performed in the temperature of 823 K, which makes possible to make a series capacity-inductance circuit to be applied to microelectronic systems.

4. CONCLUSIONS

Based on the analysis of experimental results it can be stated that for the $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposites of $x = 38,2$ at.%, produced by means of ion-beam sputtering with the application of $\text{Ar}^+\text{-O}_2^-$ ions and subsequently subdued to 15-minute isochronous annealing in the temperature of 823 K and higher, phase shift of $-90^\circ < \Theta_L < 0^\circ$ occurs within the low-frequency area, at alternating current while within the high-frequency area the phase shift is included in the value interval of $0^\circ < \Theta_H < +90^\circ$ and $\Theta_R = 0^\circ$ for the resonance frequency f_R . The observed phase-shift angle characteristics qualitatively resemble those of a conventional series RLC circuit. However, there are a few below-given differences:

- a film of nanomaterials does not include any winding that is characteristic for traditional inductance;
- within the low-frequency area in the $\Theta(f)$ characteristic a distinct minimum can be observed that does not occur in LC circuits;
- resonance frequency that characteristic for $\Theta = 0^\circ$, increases along with the temperature growth;
- low values of resonance frequency activation energy indicate that positive values of the phase-shift angle are related to the mechanism of hopping transfer of an electron whose return to the initial potential well occurs after a lapse of time τ at equally low activation energy. In that case for frequency of below $1/2\tau$ the phase lag can amount to over 2π , which means that positive phase-shift angles can occur.

Based on the performed investigations into frequency dependences of the phase-shift angle in $(\text{Co}_{45}\text{Fe}_{45}\text{Zr}_{10})_x(\text{Al}_2\text{O}_3)_{(100-x)}$ nanocomposites produced by sputtering with a combined beam of $\text{Ar}^+\text{-O}_2^-$ ions the following methods have been elaborated:

- a) a method for producing noncoil-like inductance, which consists in vapor deposition between metallic contacts of a nanocomposite film of $x = 38,2$ at.% that subsequently is subdued to 15-minute stabilizing annealing in the temperature of 848 K;
- b) a method for developing a series capacity-inductance circuit that differs from the above mentioned one by the annealing temperature, which in this case is 823 K.

REFERENCES

1. A. Saad, A.K. Fedotov, I.A. Svito, A.V. Mazanik, B.V. Andrievski, A.A. Patryn, Yu.E. Kalinin, A.V. Sitnikov, *Prog. Solid State Ch.* **34**, 139 (2006).
2. T. Kołtunowicz, *Measurement Automation and Monitoring (PAK)* **53** No11, 44 (2007).
3. A. Saad, A. Mazanik, Yu. Kalinin, J. Fedotova, A. Fedotov, S. Wrotek, A. Sitnikov, I. Svito, *Review on Advanced Materials Science* **8**, 152 (2004).
4. P. Żukowski, T. Kołtunowicz, J. Partyka, Yu.A. Fedotova, A.V. Larkin, *Vacuum* **83**, Supplement 1, S275, (2009).
5. P. Zhukowski, T.N. Kołtunowicz, J.A. Fedotova, A.V. Larkin, *Prz. Elektrotechniczn.* **86**, No7, 157 (2010).
6. P. Żukowski, T. Kołtunowicz, J. Partyka, Yu.A. Fedotova, A.V. Larkin, *Vacuum* **83** Supplement 1, S280 (2009).
7. P. Żukowski, T. Kołtunowicz, J. Partyka, P. Węgierek, M. Kolasik, A.V. Larkin, J.A. Fedotova, A.K. Fedotov, F.F. Komarov, L.A. Vlasukova, *Prz. Elektrotechniczn.* **84**, No 3, 247 (2008).
8. P. Zhukowski, J. Sidorenko, T.N. Kołtunowicz, J.A. Fedotova, A.V. Larkin, *Prz. Elektrotechniczn.* **86** No7, 296 (2010).
9. T. Kołtunowicz, *Elektronika – konstrukcje, technologie, zastosowania* **48**, No10, 37 (2007).
10. T.N. Kołtunowicz, *Measurement Automation and Monitoring (PAK)* **57** No7, 694 (2011).
11. P. Żukowski, J. Partyka, P. Węgierek, *phys. status solidi A* **159**, 509 (1997).
12. P. Zhukowski, T.N. Kołtunowicz, P. Węgierek, J.A. Fedotova, A.K. Fedotov, A.V. Larkin, *Acta Phys. Pol. A* **120** No1, 43 (2011).
13. P. Zhukowski, T.N. Kołtunowicz, P. Węgierek, A. Fedotov, J. Fedotova, A. Larkin, *Sposób wytwarzania bezuzwojeniowych indukcyjności do układów mikroelektronicznych*, Patent application no: P 390789, 22.03.2010.
14. P. Zhukowski, T.N. Kołtunowicz, P. Węgierek, A. Fedotov, J. Fedotova, A. Larkin: *Sposób wytwarzania szeregowego układu pojemność-indukcyjność do układów mikroelektronicznych*, Patent application no: P 391039, 22.04.2010.
15. A. Chochowski, *Podstawy elektrotechniki i elektroniki dla elektryków*, 169 (Warszawa: Wydawnictwo Szkolne i Pedagogiczne Spółka Akcyjna, część 2: 2009).
16. Z. Baczyński. Pat. PL 69138, Poland, publ. 31.12.1973.

Noncoil-like inductance L and a series LC circuit produced according the proposed methods can be applied to microelectronic devices.

ACKNOWLEDGEMENTS

Tomasz Norbert Kołtunowicz jest stypendystą w ramach projektu: „Nowoczesna edukacja – rozwój potencjału dydaktycznego Politechniki Lubelskiej” – Moduł II „Odnawialne źródła energii rozszerzenie i podniesienie jakości oferty edukacyjnej”. Projekt współfinansowany jest przez Unię Europejską w ramach Europejskiego Funduszu Społecznego.

