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## The influence of magnetic field on thermoelectric properties of a composite based on $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$ with a ferromagnetic filler (Co)

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The results of the study of the influence of a magnetic field on the thermal conductivity, thermoelectric power and electrical resistance of a composite based on a  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  thermoelectric (matrix) and a magnetically ordered filler (cobalt) are presented. It is shown that the introduction of a small amount of Co atoms (0.33 wt.%) into the thermoelectric matrix leads to a significant increase in the thermoelectric figure of merit  $ZT$  ( $\sim 40\%$ ). Analysis of the temperature and magnetic field dependence of the electrical resistance of the composite indicates electrical heterogeneity of the composite. The magnetic field leads to an increase in electrical resistance, a decrease in thermoelectric power and its thermal conductivity, which ultimately lead to an insignificant decrease in the thermoelectric figure of merit.

**Keywords:** thermoelectrics, magnetic field, temperature, thermoelectric figure of merit.

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### 1. Introduction

Currently, thermoelectric energy converters are used only in specific cases where other methods of energy production are difficult to implement. This is primarily due to the fact that existing thermoelectric devices are distinguished by low efficiency 6–8% and, therefore, their use is not feasible. For the thermoelectric devices to be in high demand it is required to raise significantly their efficiency which is directly related to thermoelectric Q factor  $ZT$ , expressed as  $ZT = S^2\sigma T / (k_p + k_e)$ , where  $S$  — Seebeck coefficient or thermal emf,  $\sigma$  — conductivity,  $T$  — absolute temperature,  $k_p$  and  $k_e$  — lattice and electronic components of the total thermal conductivity respectively. The product  $S^2\sigma$  is called power factor  $PF$ . Commonly used today (for example, in radioisotope thermoelectric generators), thermoelectric materials have the values  $ZT \leq 1$ . Increase of  $ZT$  to 2–3 could allow increasing the converter efficiency to the level which makes manufacture of this energy converter feasible. According to authors [1], when  $ZT = 3$ –4 the cost of energy obtained using thermoelectric converters will be comparable with the conventionally produced energy in terms of costs.

Over the past decades, researchers have paid great attention to the ways of increasing the thermoelectric converters efficiency, and several research groups have reported the achievement of  $ZT \geq 2$  in chalcogenide thermoelectric materials (see reviews [2,3]). It should be noted, however,

that these developments are still at the stage of laboratory research. The current state of research on a wide range of thermoelectric materials is regularly summarized in review papers (e.g., see [3–7]).

It follows from the expression for  $ZT$  that the thermoelectric Q factor can be increased both by increasing the power factor  $S^2\sigma$  and by reducing the thermal conductivity of the material. Yet, the parameters characterizing the efficiency of a thermoelectric material cannot be improved separately: they are interrelated and, as one parameter improves, the second one, as a rule, deteriorates. To date, a number of approaches have been proposed that can significantly reduce the lattice thermal conductivity  $k_p$  without significantly reducing the power factor  $PF$ . One of the effective methods for reducing  $k_p$  is the introduction of metallic or semiconductor nanoparticles into a thermoelectric matrix that interact effectively with phonons. Such inhomogeneities (nanoparticles) can effectively scatter phonons with their wavelengths also lying in the nanometer range, which leads to a significant decrease in  $k_p$ . This approach allowed to significantly increase  $ZT$  of some thermoelectric materials [8–10].

However, the possibilities of improving  $ZT$  by reducing thermal conductivity of a lattice are not unlimited: there are marginal minimum values of the lattice thermal conductivity for a solid body, which are determined by its structure. Therefore, in parallel, we need to look for other ways to increase  $ZT$ . As noted above, nanostructuring is one of the

most effective ways to increase the thermoelectric  $Q$  factor of materials. In this case, the  $Q$  factor rises not only due to increased scattering of phonons at the interfaces, but also due to the mechanism of the charge carriers energy filtration, leading to an increase in Seebeck coefficient. Changes in the density of the conductivity electrons near Fermi level as a result of localization of charge carriers also lead to the same result [11].

Experimental results published in a number of recent papers [12–20], indicate that magnetic interactions can have a positive effect on the thermoelectric properties of materials. Thus, in paper [12] it was demonstrated that interaction between the charge carriers and magnetic moments in chalcopyrite  $\text{CuFeS}_2$  leads to higher Seebeck coefficient  $S$  due to the growth of charge carriers effective mass, which, in its turn, results in a drastic increase of both  $PF$ , and  $ZT$ . In addition, some studies [13–20] demonstrated that if magnetically ordered inclusions are used as a filler embedded in a thermoelectric matrix, a significant increase in thermoelectric  $Q$  factor is observed. It should be noted, however, that no light has been shed on the physical mechanism to explain the increase in  $ZT$  in this case. Studies of thermoelectric properties in magnetic fields will help to clarify this issue. Due to the fact that all parameters that determine  $ZT$  (thermal conductivity, thermal emf, electrical conductivity) may depend on the magnetic field, it is natural to expect that the magnetic field will have a certain effect on  $ZT$ . A clear confirmation of this assumption is the results of [21], which showed that single-crystal samples of Bi–Sb alloy show a significant increase in  $ZT$  in relatively weak (up to 7 kOe) magnetic fields at low (nitrogen) temperatures. It should be emphasized that significant effect of the magnetic subsystem on thermoelectric properties is well known for such oxide compounds as  $\text{Na}_x\text{CoO}_2$  and  $\text{Ca}_3\text{Co}_4\text{O}_9$ . However, these studies were limited to measurements of thermal emf and electrical conductivity in magnetic fields (see reviews [22,23]). Paper [24] was an exclusion, describing the results of effect of magnetic field on the thermoelectric  $Q$  factor  $ZT$  of Cr-substituted manganites  $\text{La}_{0.65}\text{Bi}_{0.2}\text{Sr}_{0.15}\text{CoO}_3$ . It was shown that  $ZT$  strongly depends on the content of Cr, but the effect of magnetic field on  $ZT$  turned out to be weak.

Bismuth telluride and its compounds are the best low-temperature thermoelectric materials, and therefore researchers continue to focus on the related composites based on them. The purpose of this study is to find out how the magnetic field impacts the thermoelectric  $Q$  factor of a composite where  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  serves as a matrix, and ferromagnetic cobalt acts as a filler. The results of such studies will contribute to a better understanding of the physical processes that determine the thermoelectric  $Q$  factor of composites and the effect of magnetically ordered inclusions on them.

## 2. Specimens and experiment

To obtain the composite  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{ Co}$ , first the initial powders of  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  matrix and Co filler were synthesized. To synthesize powder of  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  a polyol method was used. The precursors  $\text{Bi}_2\text{O}_3$ ,  $\text{NaHSeO}_3$  and  $\text{TeO}_2$  of high purity were taken in the required stoichiometric ratio and dissolved in ethylene glycol with the addition of KOH acting as an alkaline agent. To remove the water, the resulting solution was poured into a flask and heated to boiling point with constant stirring. The flask with the solution was hermetically sealed under reflux, kept at a temperature of 458 K for 4 h and then cooled to room temperature. To separate  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  powder, the resulting suspension was centrifuged, washed with isopropyl alcohol and acetone, and then dried in a vacuum cabinet at a temperature of 373 K for 12 h. To synthesize the initial Co powder, the method of reduction from  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ .  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was used, while ethylene glycol as a coordinating agent was taken in the ratio 1:1.5 and dissolved with the addition of KOH. The mixture was heated to a temperature of 353 K until the precursors were completely dissolved and then the resulting solution was cooled to room temperature. Next, hydrazine hydrate  $\text{N}_2\text{H}_4 \cdot \text{H}_2\text{O}$  as a reducing agent was slowly introduced into the solution, the reaction mixture was heated to a temperature of 353 K and held for 6 h until the reduction of  $\text{Co}^{2+} \rightarrow \text{Co}^0$  was completed. To completely remove the impurities, the synthesized Co powder was washed with isopropyl alcohol and acetone. To obtain the bulk specimens of the studied composite, the initial powders of the matrix and filler materials, taken in the required ratio, were thoroughly mixed in a planetary mill for 30 min and consolidated by spark plasma sintering using SPS-25/10 system at a pressure of 40 MPa for 5 min and sintering temperature of 598 K. As a result cylindrical specimens with the sizes  $\varnothing 20 \text{ mm} \times 15 \text{ mm}$  were obtained. The studies of the structure of specimens  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  and  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{ Co}$  are summarized in papers [25,26].

The measurement samples consisted of plane-parallel rectangular plates  $1.5 \times 3 \times 10 \text{ mm}^3$  in size, to one of the ends of which a heater was soldered to create a temperature gradient, and the other end was in contact with a copper block as a heat sink. The heater consisted of a copper coil with a tightly wound bifilar thin constantan wire with a diameter of 0.05 mm and a resistance of 120  $\Omega$ .

Thermal conductivity was measured by the continuous heat flux method described in detail in [27]. Copper-constantan thermocouples were used as temperature sensors (wire diameter 0.05 mm). Temperature adjustment and process of thermal conductivity measurement were performed in automatic mode under program developed in the laboratory. To reduce the effect of radiation on the measurement results, the specimen was placed in a cylindrical radiation shield, the temperature of which approximately corresponds to the temperature of the specimen.

Hall coefficients  $R_x$ , concentrations of the charge carriers  $n$ , electric resistivity  $\rho$  and and charge carriers mobility  $\mu$  at  $T = 77$  and  $300$  K

	$R_x, \text{cm}^3/\text{C}$		$n, 10^{19} \text{cm}^{-3}$		$\rho, \mu\Omega \cdot \text{cm}$		$\mu, \text{cm}^2/\text{V} \cdot \text{s}$	
	77 K	300 K	77 K	300 K	77 K	300 K	77 K	300 K
$\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$	0.086	0.096	7.2	6.5	435	974	197	98
$\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33\% \text{Co}$	0.103	0.107	6	5.8	472	1034	218	103

The copper branches of the thermocouples simultaneously served to measure thermal emf and electrical resistance. The electrical resistance was determined by the four-probe method. The magnetic field up to  $80$  kOe was generated by a close-loop superconducting magnet. The temperature drop in the sample during measurements was  $4\text{--}6$  K. Vacuum was provided in the chamber during measurements  $\sim 1.33 \cdot 10^{-2}$  Pa.

According to our estimates, the electrical resistance measurement error didn't exceed  $\sim 1\%$ , thermal emf  $\sim 3\%$ , thermal conductivity  $\sim 5\%$ . During the experiments, the magnetic field was always directed perpendicular to the direction of the electric current and heat flow.

### 3. Results and discussion

First, let's discuss the results of the study of thermoelectric characteristics of composite  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{Co}$  and matrix  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$ . Composites are essentially heterogeneous systems consisting of a matrix in which filler inclusions are embedded. The structural matrix is itself a set of hexagonal plates  $\sim 500$  nm in size and  $\sim 100$  nm thick, while inclusions like „core–shell“ have characteristic sizes of  $\sim 2\text{--}8$   $\mu\text{m}$  and uniform distribution across the entire specimen volume [25,26]. Given that Co content in the matrix is  $0.33 \text{ wt.}\%$ , which is consistent with  $\sim 4 \text{ at.}\%$ , the thickness of  $\text{CoTe}_2$  shell of  $1 \mu\text{m}$  [26], and suggesting that the averaged diameter of inclusions is equal  $5 \mu\text{m}$ , we may assume that in  $\text{CoTe}_2$  compound less than half of total cobalt is contained.

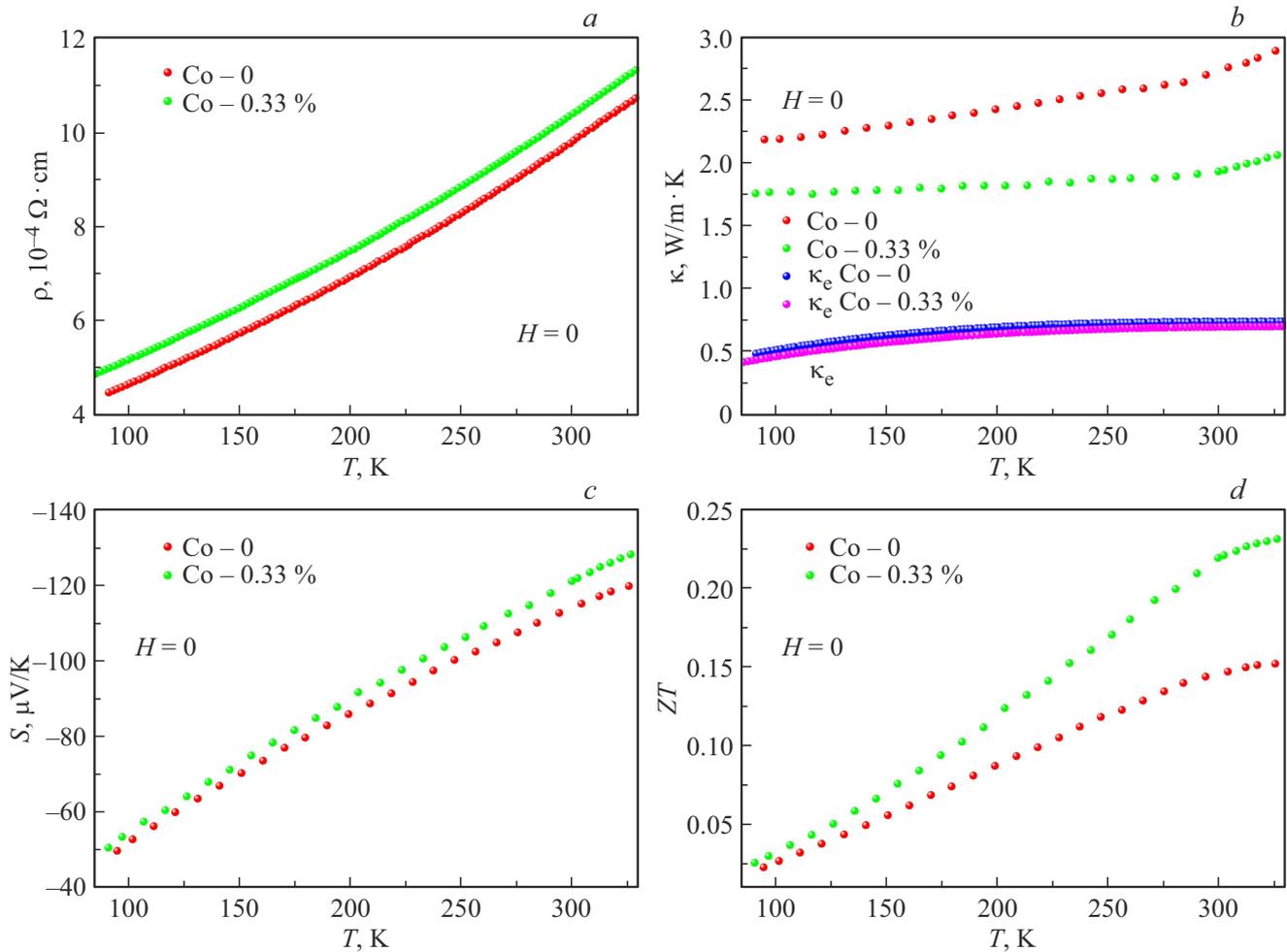
As can be seen from Figure 1, *a*, the introduction of Co inclusions into the matrix was expected to increase the electrical resistance in magnitude. Note that in case of a significant lack of Te in the composite matrix due to the formation of  $\gamma$  shell, a decrease in electrical resistance should be expected, since a lack of tellurium in  $\text{Bi}_2\text{Te}_3$  gives higher concentration of the charge carriers, and as a result, higher electrical conductivity [28,29]. The dependence  $\rho(T)$  is metallic in nature, and is explained, as in metals, by a decrease in electron mobility due to scattering by phonons, the concentration of which increases with the growth of temperature. The Hall effects measured at  $T = 77$  K and  $T = 300$  K demonstrated that concentration of the charge carriers weakly depends on the temperature variation ( $n = (6.5\text{--}7.2) \cdot 10^{19} \text{cm}^{-3}$  for  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  and  $n = (5.8\text{--}6) \cdot 10^{19} \text{cm}^{-3}$  for

$\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{Co}$ ), however, mobility of  $\mu$  is about twice less (see Table). These data confirm that the increase in resistance in these composites is associated with lower mobility of the charge carriers.

Figure 1, *b* illustrates the experimental curves  $k(T)$  for matrix  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  and composite  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{Co}$ . It also shows the electronic thermal conductivity  $k_e = L_0 T / \rho$  calculated on the basis of the Wiedemann–Franz ratio, where  $L_0 = 2.44 \cdot 10^{-8} \text{V}^2/\text{K}^2$  is the Sommerfeld value of the Lorentz number. In fact, the Lorentz number  $L$  for thermal electric materials may have somewhat lower value compared to  $L_0$ , and the actual values of  $k_e$  may differ from those shown in Figure 1, *b*, which, in fact are maximal values  $k_e$ . The validity of using the Wiedemann–Franz ratio to estimate  $k_e$  in thermoelectric materials was also discussed in the review of [1]. These possible deviations of  $k_e$  from the experimental data do not have any great impact on our line of reasoning. From Figure 1, *b* it also follows that electronic portion of thermal conductivity is a significant share of  $k_{\text{gen}}$ , reaching  $\sim 40\%$  for a composite and  $\sim 30\%$  for matrix.

A significant decrease in thermal conductivity in composite Co– $0.33\%$  compared to the initial sample Co– $0$  may be due to several factors: 1) more intensive phonon scattering processes on inclusions of Co/ $\text{CoTe}_2$  and 2) due to phonon scattering on vacancies of tellurium, which are formed during the preparation of the composite. Authors of [30] indicated the significant role of vacancies in limiting the thermal conductivity of thermoelectric materials. We assume that a small amount of filler introduced ( $0.33 \text{ wt.}\% \text{Co}$ ) does not create a sufficiently high concentration of vacancies creating a noticeable effect on heat transfer in the composite and the main reason for lower thermal conductivity in the composite Co– $0.33\%$  is the scattering of phonons on Co/ $\text{CoTe}_2$  inclusions.

Nano-structuring can simultaneously with a decrease in phonon thermal conductivity lead to an increase in Seebeck coefficient due to the effect of electron energy filtration, which means a higher proportion of the charge carriers with their average energy exceeding Fermi energy. Figure 1, *c* shows the curve of thermal emf versus temperature for both samples, where we may see that introduction of small-dimensional ferromagnetic particles into the matrix leads to higher  $S$ , which is most evident in the near-room temperature.



**Figure 1.** Electrical resistance (a), thermal conductivity (b), thermo-emf (c) and thermoelectric Q factor (d) versus temperature for the matrix  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  (Co–0) and the composite  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33$  wt.% Co (Co–0.33 %).

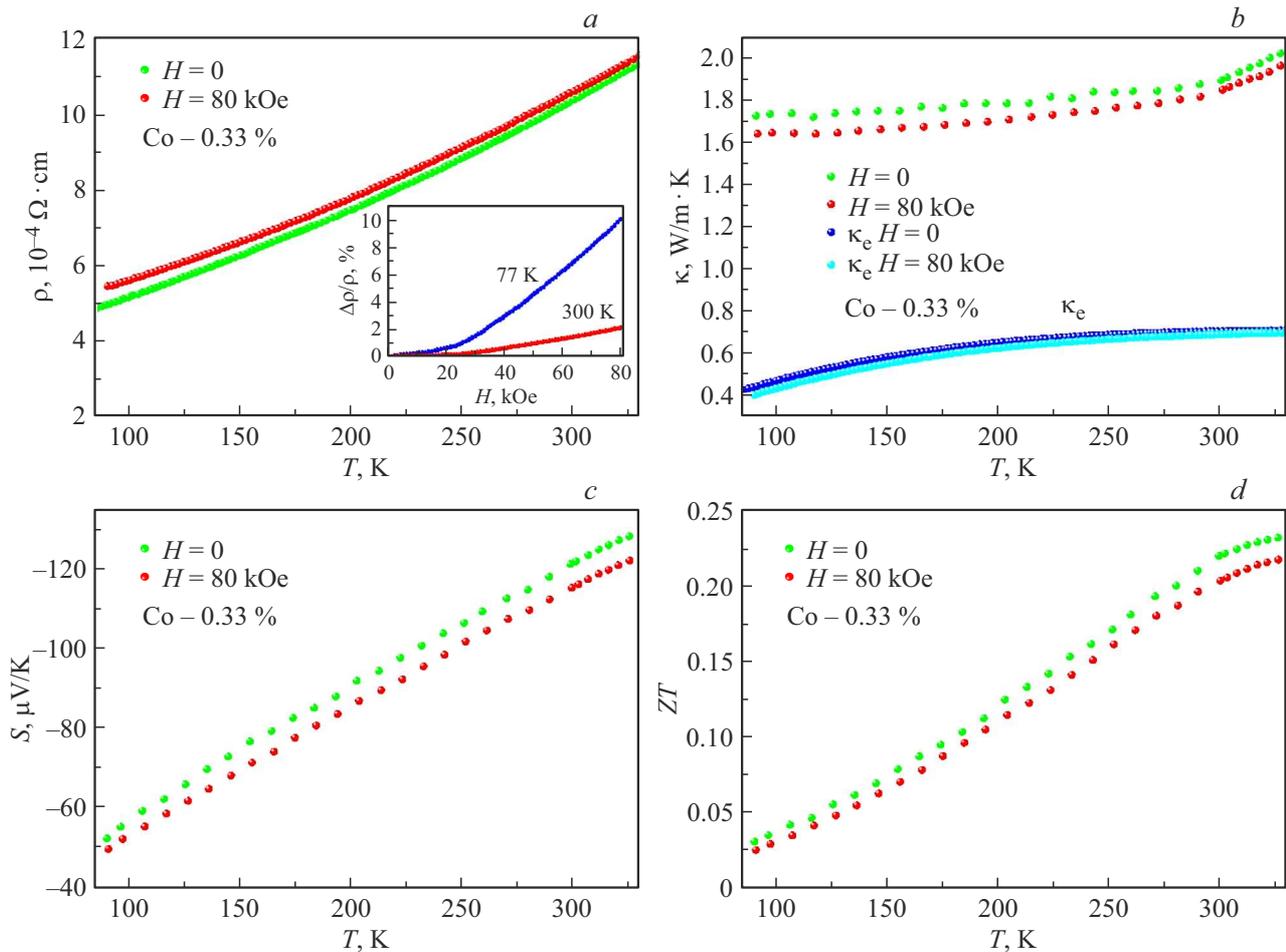
And, finally, Figure 1, *d* shows the thermoelectric Q factor versus temperature for the both specimens. The combined effect of the three parameters that determine  $ZT$  and affect the quality factor in different ways leads to a significant increase in  $ZT$  compared to a matrix without ferromagnetic inclusions, although the values themselves  $ZT$  are not very high: 0.14 and 0.23, respectively.

Let's discuss the findings on potential effect of magnetic field on the composite's thermoelectric Q factor.  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33$  wt.% Co and matrix  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$ . It was shown above that the introduction of magnetic-ordered inclusions into the matrix leads to a significant growth of  $ZT$  of the composite. Also, let's consider how magnetic field impacts  $ZT$  of the composite. The results of these studies are shown in Figure 2.

As can be seen from Figure 2, *a*, a positive magnetoresistive effect is observed across the studied temperature range. In general, this is the expected result: a magnetic field should act on a moving electric charge. More interesting is the magnetic field dependence of resistance  $\Delta\rho/\rho_0(H)$ , shown in Figure 2, *a* (insert) for two temperatures. In the

study [31] it was shown that when Ni atoms are introduced in the matrix of thermoelectric  $\text{Bi}_2\text{Te}_3$  it results in a strong magnetic inhomogeneity of the specimen, corresponding to Parish–Littlewood model [32] (alternating regions with high and low conductivity): positive magnetoresistive effect, a crossover of  $\Delta\rho/\rho_0(H)$  ratio from the square to a linear one with the growth of  $H$ , decrease of  $\Delta\rho/\rho_0$  with the temperature growth. In this case, we see a similar temperature-dependent behavior of  $\Delta\rho/\rho_0(H)$ , which may also indicate an electrical heterogeneity of the composite.

Figure 2, *b* illustrates how magnetic field impacts the thermal conductivity of a composite. The magnetic field does not directly affect the phonon component of thermal conductivity, and, obviously, the changes in  $k$  are associated with the manifestation of Maggi–Righi–Leduc effect — reduction of the electronic component of thermal conductivity under the influence of magnetic field. The change in electronic thermal conductivity  $L_0T(1/\rho_0 - 1/\rho_H)$  and the experimental values of thermal conductivity in a magnetic field of 80 kOe and without magnetic field as estimated from the Wiedemann–Franz ratio are comparable values



**Figure 2.** Dependence diagram  $\rho(T)$ ,  $k(T)$ ,  $S(T)$  and  $ZT(T)$  for the composite  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9} + 0.33 \text{ wt.}\% \text{Co}$  in magnetic field of 80 kOe and without field. The insert window in Figure 2, *a* shows the curves of magnetoresistance  $\Delta\rho/\rho$  versus magnetic field measured at  $T = 77$  and 300 K.

and correspond to existing ideas about the nature of the magnetic field influence on electronic thermal conductivity.

Figure 2, *c* gives a diagram of thermal emf versus temperature in magnetic field and without magnetic field. The sign  $S$  indicates the electronic nature of current carriers, and the dependence  $S(T)$  corresponds to the general notion of how the diffusion thermal emf depends on temperature, according to which  $S$  increases linearly with  $T$ . Under the influence of magnetic field, thermal emf slightly varies (decreases in absolute value).

The dependence of  $ZT$  on temperature, based on the above experimental data, is shown in Figure 2, *d*. It can be seen from the figure that magnetic field leads to a slightly lower  $ZT$ , which is most likely the result of the prevailing influence of  $S$  on  $ZT$  in the magnetic field. Magnetic field impacts  $ZT$  of  $\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  matrix in almost the same way.

#### 4. Conclusion

The thermal conductivity, thermal emf, and electrical resistance of a composite based on a thermoelectric material

$\text{Bi}_2\text{Te}_{2.1}\text{Se}_{0.9}$  and a magnetic-ordered filler (cobalt) were measured. It is shown that introduction of Co atoms into the matrix of a thermoelectric material leads to significant changes in the measured properties: higher electrical resistance, higher thermal emf, and much lower thermal conductivity which is mainly due to the appearance of additional scattering centers. The result of such changes is a significant increase in the thermoelectric  $Q$  factor by about  $\sim 40\%$ . Estimates show that the electronic contribution to thermal conductivity is a significant proportion ( $\sim 30\%$  for the matrix and  $\sim 40\%$  — for the composite).

Magnetic field has a significant effect on the measured properties: It causes an increase in electrical resistance, a decrease in thermal emf and thermal conductivity. The result of this effect is a slight decrease in the thermoelectric  $Q$  factor.

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## Conflict of interest

The authors declare that they have no conflict of interest.

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