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Nernst-Ettingshausen coefficient and charge-carrier mobility in the $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ system

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The paper presents the results of the study and analysis of the temperature dependences of the Nernst-Ettingshausen coefficient, Q , in samples of the $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ system with different fixed cobalt content ($z = 0.25, 0.4$) and varied calcium content ($x = 0-0.2$). Using the values of the energy spectrum parameters in the studied samples, obtained earlier from the thermopower analysis, we performed a quantitative analysis of the experimental data and determined the values of the electron mobility and the degree of the dispersion law asymmetry. The influence of calcium on both the experimental $Q(T = 300\text{ K})$ value, and the values of the calculated charge-carrier parameters was observed to depend on the content of cobalt ions in the lattice. It is shown that both the type of dependence of these parameters on the calcium content and the differences in these dependences for the two studied sample series are caused by the peculiarities of the transformation of the $YBa_2Cu_3O_y$ energy spectrum under the influence of each of the studied impurities.

Keywords: high-temperature superconductors, doping, thermopower, energy spectrum.

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1. Introduction

Oxide high-temperature superconductors (HTSC) have been intensively studied for almost forty years, since the discovery of compounds of this class in 1986. Despite a huge amount of data on various properties of HTSC materials, as well as some progress in their practical use, many fundamental questions of the physics of these compounds, primarily the establishment of the mechanism of electron pairing, remain unresolved.

The electron system in HTSC materials is characterized by the presence of strong correlation effects, which makes it impossible to use well-known classical single-electron models for quantitative analysis of the results of various experiments. One of the examples confirming this is the development of a large number of new approaches (both theoretical, phenomenological, and even simply descriptive) capable of explaining the features of electronic transport in oxide HTSC in a normal state. It is well known that the experimentally observed temperature and concentration dependences of all kinetic coefficients (especially their combination) are a distinctive feature of HTSC materials [1–5]. Numerous attempts to describe them both qualitatively and quantitatively (see, for example, [6–16]) do not allow definitively determining, as in the case of many other materials, the values of such important parameters of the electronic system as, for example, the concentration and mobility of charge carriers, the value of the density function

of states at the level of Fermi, the mechanism of conduction, and others.

We have also proposed an electronic transport model — a narrow band model [17]. Despite the fact that this model is phenomenological, it can be successfully used to obtain quantitative information about the parameters of the normal state in HTSC materials. Initially, the analysis of temperature dependences of the thermopower S was used for this purpose. Based on this analysis, the main general features of the structure of the cuprate HTSC energy spectrum, the mechanism of its change under the action of various types of alloying, as well as the peculiarities of the influence of some of the alloying impurities on its parameters were determined [4,17–22]. However, later we were able to expand this model and apply it also to describe the Nernst-Ettingshausen coefficient (NEC, Q) [23], which additionally allows obtaining values of the mobility of charge carriers for this coefficient from the analysis of experimental data [5,24]. It should be noted that attempts to analyze this kinetic coefficient in a wide range of temperatures above the critical temperature range, T_c , presented in the literature, are extremely scarce and are rather purely qualitative in nature [25–28].

This study is devoted to the application of the approach developed by us, based on a joint analysis of the temperature dependences of the thermopower and NEC, to the yttrium HTSC system with double substitutions — $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$. It is well known

that calcium, suppressing superconductivity in optimally doped $\text{YBa}_2\text{Cu}_3\text{O}_y$ [18,21,29,30], leads to its restoration when nonstoichiometric composition is introduced into samples, and in cases of both the presence of oxygen deficiency [18,21,31], as well as pre-introduced other impurities [21], in particular, cobalt in the position of chain copper [21,32,33]. The aim of the paper was to study the characteristics of the temperature and concentration dependences of NEC in samples with a fixed cobalt content and a variable calcium content, to conduct their quantitative analysis and to clarify the nature of the effect of calcium on the mobility of charge carriers.

2. Samples and methodology

Experimental studies were carried out on ceramic samples of the compositions $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$ at two different fixed cobalt contents ($z = 0.25$ and $z = 0.4$) and varying calcium content ($x = 0.0, 0.05, 0.1, 0.2$). A standard tristadium solid-phase synthesis procedure was used for their manufacture with a sequential increase in the annealing temperature of the mixture (from 900 to 930 °C) and intermediate grindings. After the synthesis was completed, all samples were jointly annealed in an air atmosphere in flowing oxygen at $T = 450$ °C for 6 h in order to achieve their optimal oxygen composition. The uniformity and single-phase character of all samples was confirmed by measuring the local values of the thermopower at various points on their surfaces and X-ray phase analysis, respectively.

Previously, the temperature dependences of resistivity and thermopower were measured on all the samples studied in this work; detailed data on the results obtained in this case are given in Ref. [21]. We only note here that the value of the critical temperature T_c increases sequentially with the increase of the calcium content in both studied systems, from 60.2 to 78.6 K for the series with $z = 0.25$ and from 32.0 to 72.6 K for the series with $z = 0.4$, which confirms the presence of the effect of superconductivity reduction during calcium doping.

For all samples, the temperature dependences of the NEC were measured in the temperature range of $T = 100$ –300 K. The value of the induction of the measuring magnetic field was $B = 1.8$ T, the value of the temperature difference applied to the sample was $\Delta T = 10$ –15 K. Since the values of NEC in HTSC materials in the normal phase are extremely small [5], thin (about 1 mm) in the direction of the temperature drop and long (about 10 mm) in the direction of the voltage were used to increase the measured signal. The measurements were conducted on an original experimental setup developed in the laboratory using a fully automated technique that allows programmatically compensating for the values of all spurious signals that occur during the measurement of the Nernst-Ettingshausen signal [34,35], and thereby achieving high accuracy in measuring the values of the NEC.

3. Experimental results and their discussion

The experimental temperature dependences of the NEC for samples of both studied systems with different cobalt admixture contents are shown in Figures 1 and 2, respectively. In general, the dependencies $Q(T)$ demonstrate features characteristic of other alloyed samples of the HTSC yttrium system [5]. At low temperatures ($T \approx 100$ K), the values of Q are extremely small; they increase quite sharply with the increase of the temperature, and they slightly change in the high temperature range (at $T > 200$ –250 K, depending on the specific composition of the sample). However, it is necessary to note some features of the effect of calcium admixture on the behavior of NEC.

Figure 3 shows the dependences of the NEC value at $T = 300$ K, $Q_{300\text{K}}$, on the calcium content for both series of studied samples of the compositions $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$. Here, for comparison, similar dependences of the values of the thermopower at room temperature, $S_{300\text{K}}$, are shown according to Ref. [21]. The value of $Q_{300\text{K}}$ for the starting (calcium-free) sample of the series with $z = 0.4$ is approximately two times higher than in the case of the series with $z = 0.25$, which corresponds to the previously discovered nature of the effect of a single substitution of copper with cobalt [23]. At the same time, the effect of the additional injected calcium on this value depends on the cobalt content in the samples. The value of $Q_{300\text{K}}$ consistently decreases with a large value ($z = 0.4$) with an increase of x , whereas it almost does not change with a smaller value ($z = 0.25$).

It should be noted that in the case of single calcium doping (samples with the composition $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_3\text{O}_y$), the value of $Q_{300\text{K}}$ hardly changes with increase of x [21,23]. In addition, an increase in calcium in the system of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$ leads, as previously indicated, to a strong increase in the value of the critical temperature, as well as to a sharp drop in the values of resistivity and the thermopower and Hall coefficients [21]. In particular, as can be seen from Figure 3, the value of $S_{300\text{K}}$ in the range of $x = 0.0$ –0.2 decreases from 89 to 25 $\mu\text{V/K}$ at $z = 0.4$ and from 33 to 8 $\mu\text{V/K}$ at $z = 0.25$, i.e., 3.5–4 times, whereas the value of the NEC decreases only by about 2 times even in the series where its drop is observed ($z = 0.4$). These facts indicate that the change in the values of $Q_{300\text{K}}$ is not caused by a change in the charge balance in the lattice $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$ with the introduction of Ca ions into it and the compensating (compared with the influence of Co) effect of this impurity on it, as in the case of the thermopower [21,33], and with changes in other parameters of the studied systems under the action of calcium doping. Thus, to explain the results obtained, it is necessary to analyze them within the framework of the model used in order to determine the values of the parameters of the charge carrier system in samples with different calcium contents.

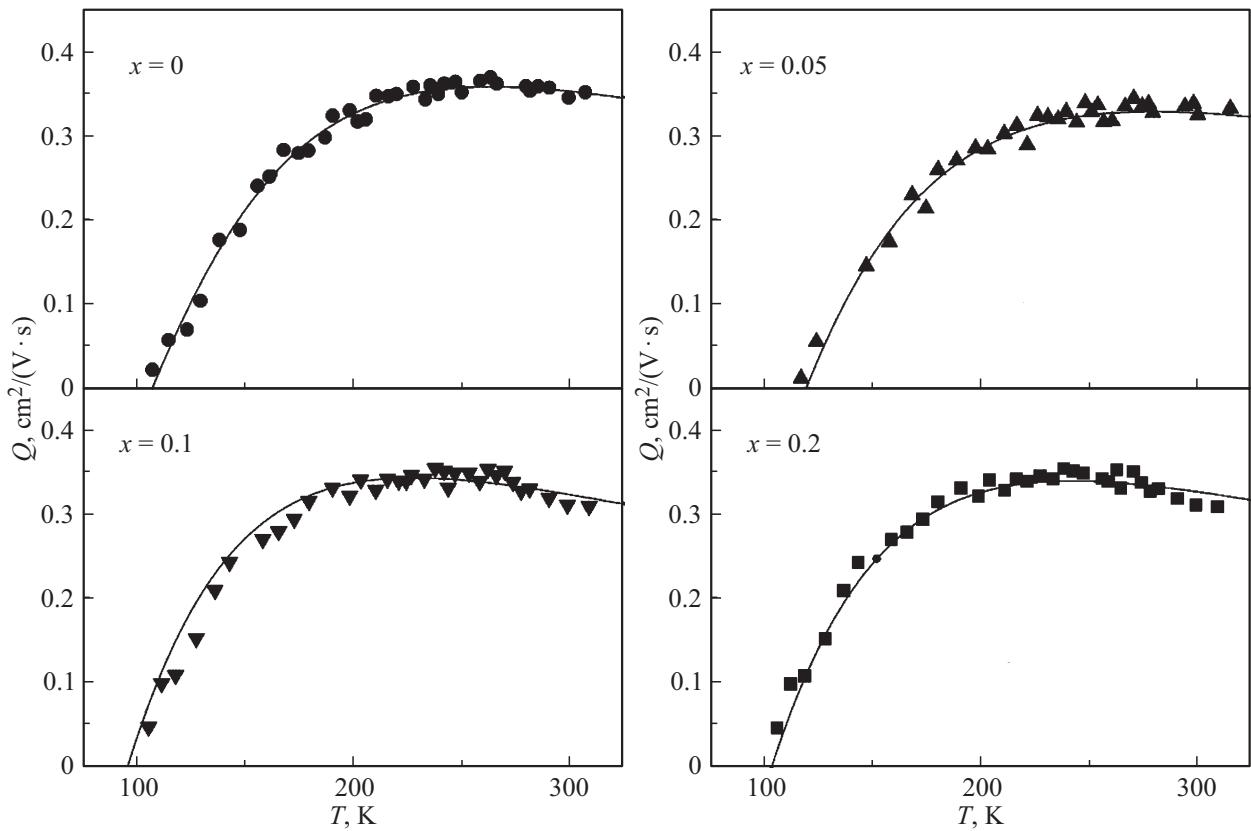


Figure 1. Nernst-Ettingshausen coefficient in samples $Y_{1-x}Ca_xBa_2Cu_{2.75}Co_{0.25}O_y$. Symbols — experiment, lines — calculation based on the narrow band model.

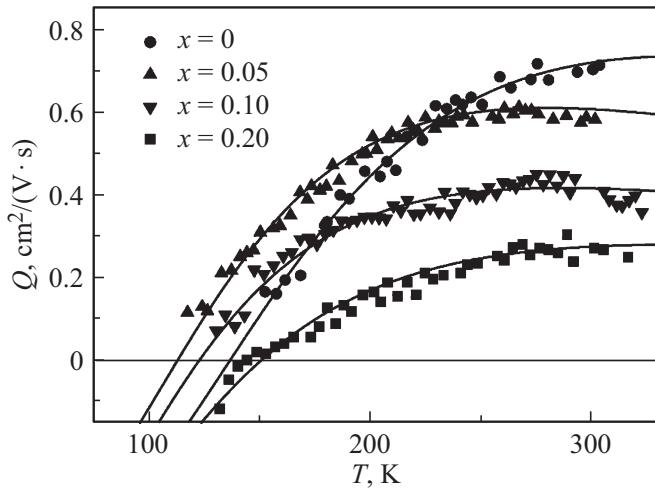


Figure 2. Nernst-Ettingshausen coefficient in samples $Y_{1-x}Ca_xBa_2Cu_{2.6}Co_{0.4}O_y$. Symbols — experiment, lines — calculation based on the narrow band model.

The analysis of the obtained results was carried out using the previously developed narrow band model [17,23]. The most detailed description of the main provisions of this model is given in Ref. [4], and the features and method of its use for the analysis of temperature dependences of NEC are

given in Ref. [5]. Here we will only briefly point out that, assuming that there is a narrow peak of the density function of states in the energy spectrum of the HTSC, in which the Fermi level is located, it is possible, using expressions for kinetic integrals obtained from solving the kinetic Boltzmann equation in the approximation of relaxation time, to obtain analytical expressions for the temperature dependences of all the main kinetic coefficients. At the same time, the values of the thermopower and NEC coefficients can be calculated with accuracy to absolute values, which makes it possible to use the formulas obtained for the dependencies $S(T)$ and $Q(T)$ for quantitative analysis of experimental data. According to Ref. [23], the value of the NEC in the framework of the narrow zone model can be calculated as follows:

$$Q = -\frac{1}{eT} u \left(-\frac{I_{\sigma_1} I_{\sigma_{H_0}}}{I_{\sigma_0}^2} + \frac{I_{\sigma_{H_1}}}{I_{\sigma_0}} \right), \quad (1)$$

where

$$I_{\sigma_i} = \int_{-\frac{W_{\sigma}}{2} + bW_D}^{\frac{W_{\sigma}}{2} + bW_D} \varepsilon^i \left(-\frac{\partial f_0}{\partial \varepsilon} \right) d\varepsilon,$$

$$I_{\sigma_{H_i}} = \int_{-\frac{W_{\sigma}}{2} + kW_D}^{\frac{W_{\sigma}}{2} + kW_D} \varepsilon^i \left(-\frac{\partial f_0}{\partial \varepsilon} \right) \text{sign}(\varepsilon - kW_D) d\varepsilon,$$

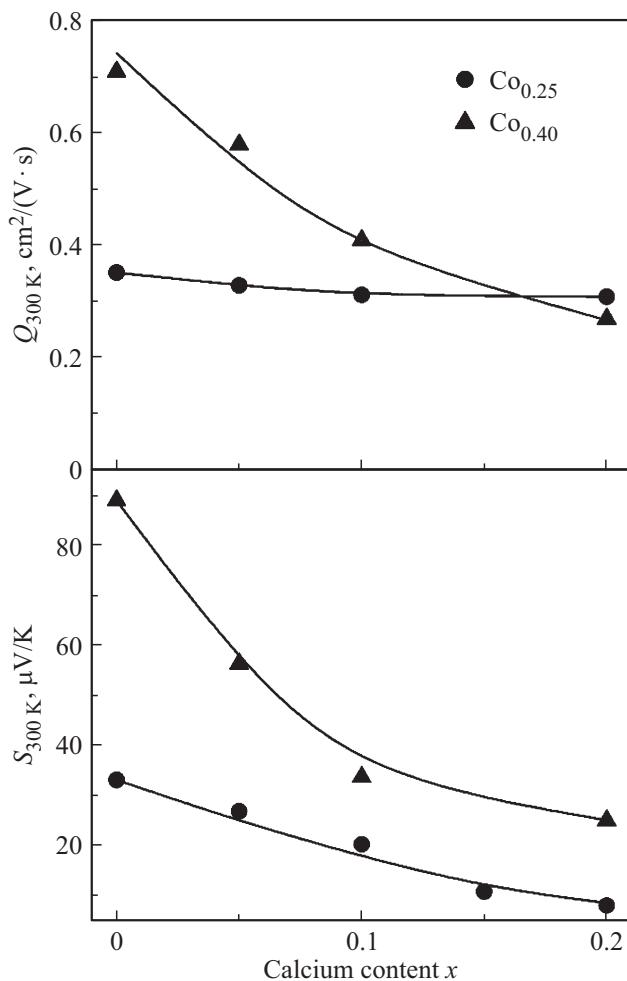


Figure 3. Changes in the values of the thermopower and Nernst-Ettingshausen coefficients at $T = 300\text{ K}$ in samples of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$.

the origin of the energy in the integrals corresponds to the middle of the band, e — the electron charge, f_0 — the equilibrium Fermi-Dirac distribution function, and the electrochemical potential μ is calculated using the formula

$$\mu = k_B T \cdot \ln \frac{\sinh(FW_D(2k_B T))}{\sinh((1-F)W_D/(2k_B T))} - 2bW_D$$

(k_B is the Boltzmann constant).

The above expressions include six model parameters: F is the degree of electron filling of the band; W_D and W_σ is the total effective width of the conducting band and the effective width of the range of conducting states, the ratio of these values $C = W_\sigma/W_D$ characterizes the proportion of delocalized states in the band; b is the degree of asymmetry of the band; k is the degree of asymmetry of the dispersion law; u is the value of electron mobility averaged over the entire band. Obviously, it is impossible to use the expression (1) to determine the values of all model parameters based on experimental dependencies $Q(T)$. However, if the temperature dependences of both the NEC

and the thermopower are measured for a sample of a given composition, then the first four parameters can be determined from the analysis of the dependencies $S(T)$, and then simply substituted into the expression (1), which makes it possible to unambiguously determine the values of k and u based on the dependencies $Q(T)$.

The results of the thermopower analysis and their detailed discussion are presented in Ref. [21]. Figure 4 shows the data obtained in Ref. [21] about changes in the total effective band width and the degree of delocalization of band states, which will be necessary in the future when interpreting the results on changes in electron mobility with an increase in the doping level obtained from the NEC analysis. As for the other two parameters — F and b — the degree of electron filling of the zone in both series of samples of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$ decreases sequentially with the increase of x , which is caused by the effect of charge compensation of the effect of the Co impurity by the introduced Ca ions [21]. As a result, a decrease in the value of W_D and an approximation of the degree of filling of the zone to $F \approx 0.5$ (which means a shift of the Fermi level to the middle of the band) lead to an increase in the value of the density function of states at the Fermi level, which is the main reason for the observed increase in the value of the critical temperature. The degree of asymmetry of the band increases linearly with the growth of x according to the universal law for all calcium-doped samples of the $\text{YBa}_2\text{Cu}_3\text{O}_y$ $b \approx -0.12x$, which is a consequence of the formation of an additional peak of calcium states in the energy spectrum [21].

The values of the model parameters found in Ref. [21] from the analysis of dependency $S(T)$ were used in modeling the temperature dependences of the NEC using the formula (1). The calculated curves corresponding to the optimal values of k and u , at which the best agreement of these curves with experimental data is achieved, are shown in Figures 1 and 2. The values of the electron mobility in the studied series of samples obtained from the results of

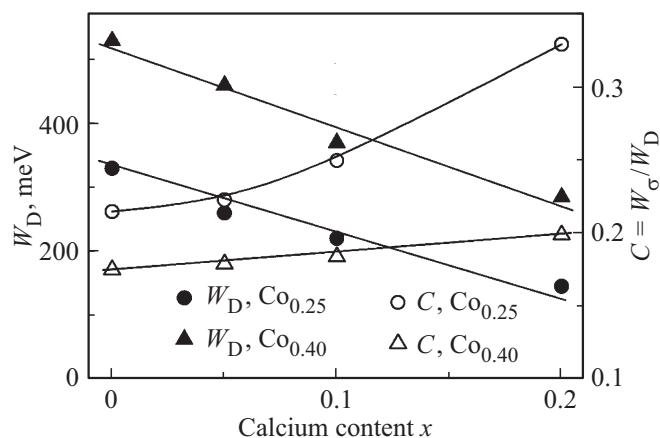


Figure 4. Dependence of the effective band width and the degree of delocalization of states on the Ca content in samples of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$.

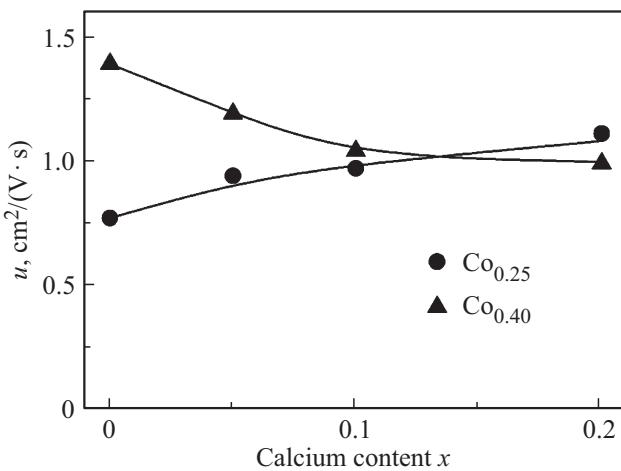


Figure 5. Concentration dependences of electron mobility in samples of $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$.

the analysis are shown in Figure 5. As in the case of other alloyed HTSC of the yttrium system [5], the mobility in all samples of the system of $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ has very low values.

It can be seen from Figure 5 that the effect of calcium on the mobility value significantly depends on the level of doping of the samples with cobalt. The mobility slightly increases at $z = 0.25$ with the increase of the doping levels, and it decreases at $z = 0.4$. It should be noted that the values shown in Figure 5 are not the so-called Nernst mobility, defined as $Q/(k_B/e)$ and varying in accordance with the change in values of Q (see Figure 3). The value of u in the framework of the narrow band model is defined, as in the classical case, as $u = e\langle\tau\rangle/m^*$, where $\langle\tau\rangle$ is the average relaxation time of charge carriers, m^* is their effective mass [5,23]. The only difference is that in the presence of a narrow band, the relaxation time should be averaged over the entire band and with a weighting factor of $(-\partial f_0/\partial\epsilon)\epsilon$, and not $(-\partial f_0/\partial\epsilon)\epsilon^{3/2}$, as in the classical theory of kinetic phenomena [5].

As mentioned above, it is necessary to use data on changes in other parameters of the charge carrier system to explain the obtained dependencies $u(x)$. As can be seen from Figure 4, an increase in the Ca content in $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ causes a significant narrowing of the band (a decrease in the values of W_D) and, at the same time, an increase in the proportion of delocalized states (an increase in the values of C). The physical reason for this is that non-covalent substitutions in $YBa_2Cu_3O_y$ lead, with an increase in their content, to an increase in the degree of disordering in the system — both due to the direct substitution of lattice ions and, to a greater extent, by increasing the oxygen content (if the valence of the impurity atoms is greater than the valence of the corresponding atom in the lattice, as in the case of substitution of $Co \rightarrow Cu$). This leads to the implementation of the mechanism of Anderson localization of states [36] and corresponding

changes in the structure of the band [4]. As a result, in the starting (without calcium) samples of both series $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ the band is quite wide compared to optimally doped $YBa_2Cu_3O_y$, and a significant part of the band states are localized (see Figure 4). Calcium, due to its lower valence compared to yttrium, has the opposite effect on the oxygen content, thereby leading to an increase in the degree of ordering of the system and the inverse dependence of the above parameters [21]. The resulting narrowing of the band leads to an increase in the effective mass of electrons, and a decrease in the degree of defects in the oxygen subsystem leads to an increase in their relaxation time. The first should cause a decrease in the value of mobility, and the second, on the contrary, should increase it.

As can be seen from Figure 4, with an increase in the calcium content in both series of samples $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ the width of the band W_D decreases sequentially with an increase of x , while the slope of the dependence $W_D(x)$ is the same for these two series of samples. The degree of delocalization of states increases in both series, however, to varying degrees. In the case of $z = 0.4$, this growth is rather weak, so the effect of narrowing the band and, consequently, increasing the effective mass of electrons on the mobility value is more significant, which leads to its decrease (see Figure 5). In the series of samples with $z = 0.25$, there is a significantly stronger increase in the values of C . As a result, the effect of increasing the relaxation time of electrons on their mobility is more significant than the effect of narrowing of the band, which leads to an increase in its values. The difference in the degree of increase in the values of C in the two studied series of samples, leading to a change in the nature of the dependence of $u(x)$, can be explained taking into account data on the positions occupied by cobalt in the lattice of doped $YBa_2Cu_{3-z}Co_zO_y$. It is known that at low concentrations ($z < 0.2-0.3$), all Co ions are located at copper positions in CuO chains [37,38]. In this case, the effect of this impurity on the properties of the compound is almost solely due to its effect on the state of the oxygen subsystem (an increase in oxygen content and an increase in the disorder in the distribution of its atoms in various positions). It is this effect that is compensated by the additional Ca ions introduced into the lattice, moreover, as shown by the results of a study of the thermopower in the system of $Y_{1-x}Ca_xBa_2Cu_{3-x}Co_xO_y$, with the same amount of Ca and Co impurities introduced, this compensation is almost complete [39]. For this reason, for a series of samples with $z = 0.25$, there is a strong increase in the degree of ordering with an increase in the Ca content. At high concentrations of cobalt, some of its atoms are located in copper positions in the CuO_2 planes [37,38] responsible for the formation of the conducting band in $YBa_2Cu_3O_y$, and it has a direct effect on the disorder in their electronic structure. This effect cannot be compensated by the influence of calcium ions, which leads to a weaker increase in C values in a series of samples with $z = 0.4$.

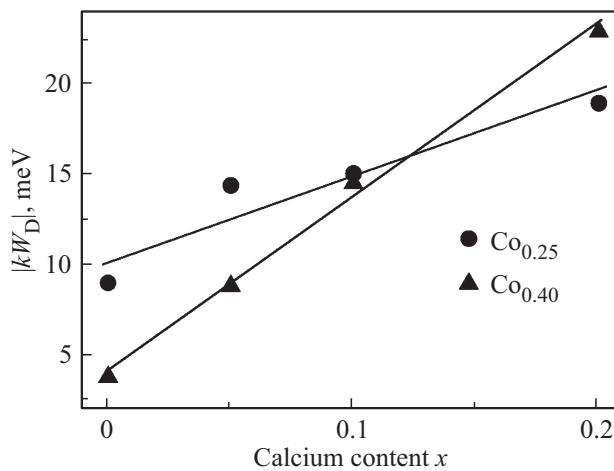


Figure 6. Effect of calcium doping on the position of the Hall conductivity change point in samples of $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$.

The values of the parameter k , found from the dependence analysis $Q(T)$ and characterizing the degree of asymmetry of the dispersion law, are negative, as for all doped samples of the system of $YBa_2Cu_3O_y$ [5]. As the Ca content increases, they increase in absolute value from 0.027 to 0.13 for the series with $z = 0.25$ and from 0.007 to 0.08 for the series with $z = 0.25$. According to the narrow band model used by us for the analysis, the asymmetry of the dispersion law is modeled by shifting the point of change of sign of the Hall conductivity from the middle of the band by kW_D [5,23]. The obtained values of $|kW_D|$ for all samples are shown in Figure 6.

In general, the data obtained correspond to the nature of the influence of single impurities of Co and Ca on the asymmetry of the dispersion law. As shown by previously obtained results in Ref. [5], the presence of asymmetry of the law of dispersion is characteristic of samples of the $YBa_2Cu_3O_y$ system of any composition including the optimal one. In this case, its degree changes in case of doping with those impurities that directly affect the structure of the conductive zone. The value of $|kW_D|$ begins to decrease at $z > 0.2$ in the $YBa_2Cu_{3-z}Co_zO_y$ system, i.e. when Co atoms begin to partially replace planar copper (see above), resulting in samples of $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ in the absence of Ca ($x = 0$) at $z = 0.4$, this value is lower than at $z = 0.25$. An increase in the Ca content in samples of $YBa_2Cu_3O_y$ system of any cationic composition leads to an increase in the value of $|kW_D|$, which is associated with the appearance of an additional peak of calcium states in the energy structure of the conducting band [21]. This is also observed in the case of the systems studied in this paper (see Figure 6). At the same time, this growth is stronger in the series with $z = 0.4$, since the calcium peak is formed against the background of a band partially destroyed by the presence of Co ions in the positions of planar copper, which enhances the degree of influence of an increase in this peak on the asymmetry of the dispersion law.

4. Conclusion

An experimental study and analysis of the temperature dependences of the Nernst-Ettingshausen coefficient in samples of $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ with different fixed Co content and variable Ca content. The following main results were obtained.

1. The temperature dependences of the NEC in the studied systems demonstrate typical features of doped yttrium HTSC, however, the nature of the effect of Ca on their appearance, as well as the absolute values of Q at room temperature, significantly depend on the content of Co ions in the lattice.

2. It is shown that the use of a narrow band model for the analysis of electronic transport in the system $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ makes it possible to simultaneously quantify the temperature dependences of the thermopower and NEC coefficients and, from the analysis of the latter, in addition to the main parameters of the charge carrier system, determine the values of electron mobility and the degree of asymmetry of the dispersion law.

3. The value of electron mobility in both series of samples changes rather weakly with the increase of Ca content near $u \approx 1 \text{ cm}^2/(\text{V} \cdot \text{s})$, although in different ways — the mobility increases with a low Co content, and it decreases with a high Co content.

4. An increase in the Ca content in both systems leads to an increase in the asymmetry of the dispersion law, which is caused by the introduction of additional states by calcium into the zone, while with a higher Co content this increase occurs more strongly than with a lower one.

5. All the discovered features of the concentration dependences of mobility and the degree of asymmetry of the dispersion law in both studied series of samples, including the differences between them, can be explained taking into account the effects of Ca and Co on the structure and parameters of the band responsible for conductivity in the $YBa_2Cu_3O_y$ system in the normal phase.

Conflict of interest

The authors declare no conflict of interest.

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