=== **THERMOPHYSICAL PROPERTIES OF MATERIALS** ===

Thermophysical Properties of Solid Yttrium−Holmium Solutions in the Temperature Range from Room Temperature to 1400 K

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Abstract—The thermophysical properties (thermal diffusivity, specific heat, and thermal conductivity) and resistance of solid solutions (alloys) of rare-earth metals (yttrium and holmium) have been experimentally studied. It is established that, on the whole, these properties obey the same laws as pure rare-earth metals. Heat transfer in Y−Ho alloys at the considered temperatures is mainly performed by electrons as well. The existing quantitative differences between the properties are due to the different influences of the different mechanisms of carrier (electron) scattering. A procedure to separate the contributions to electron scattering based on the Mott model is considered. The roles of phonon-, magnetic-, and impurity-scattering mechanisms are determined. It is established that the magnetic-scattering intensity monotonically decreases upon heating and with a decrease in the holmium concentration. Nordheim's rule is valid for impurity scattering, which indicates stability of the energy spectrum structure of collective alloy electrons.

INTRODUCTION

Rare-earth metals (REMs) are condensed phases of some elements from the Mendeleev periodic table: scandium, yttrium, and lanthanides [1−5]. Many physical and chemical properties of REMs are similar; however, some characteristics (such as molar mass and ion magnetic moment) may differ significantly. Thus, REMs are the only group of elements for which their alloys can be used to simulate a wide range of various physical situations and, thus, to trace the effects of particular characteristics of components on the resulting properties.

To date, many thermophysical properties of pure REMs have been studied [1−7]. The properties of alloys have been mainly analyzed below room temperature, while the characteristics of high-temperature alloy are barely studied. Previous works on the thermal diffusivity of yttrium−holmium (Y−Ho) alloys demonstrated that its behavior is peculiar in many aspects [8]. This study continues the previous research.

It should be noted that the range of application of REMs in industry (especially in electronics and machine building) have sharply increased in recent years. This has stimulated research on REMs and their alloys, because the results are of both scientific and practical interest.

SAMPLE CHARACTERISTICS

In this study, we analyzed the thermophysical properties of Y−Ho alloys. The properties of the components (yttrium and holmium) can be assumed to be reliably determined [1−7]. Both metals belong to the so-called yttrium REM subgroup. This means that they have similar energy structures of collective electrons [9−11], identical crystal structures, and close lattice parameters [1−5]. Thus, these metals in alloys form a continuous series of solid solutions [12]. The latter circumstance makes it possible to analyze the physical properties of these alloys from a unified point of view, when it is taken into account that a change in the concentration causes only corresponding quantitative changes in the properties.

Despite the similarity of many of the above-mentioned characteristics, the masses of yttrium and holmium ions differ significantly (by a factor of almost 2). Another important difference concerns their magnetic properties [13]. Yttrium is a Pauli paramagnet; therefore, its trivalent ion has no magnetic moment. A trivalent holmium ion has the largest (among REMs) total quantum number and thus possesses significant magnetism. This ion is naturally a source of magnetic carrier scattering. As a result, the carrier scattering in these alloys is caused by all main mechanisms at high temperatures (specifically, phonon, impurity, and magnetic mechanisms).

The initial materials for the preparation of alloys (yttrium and holmium) were purified via vacuum dis-

tillation. Mass spectrometry revealed that the total impurity content was 0.04% for each of the components. The ratio of resistances was 20 at room temperature and at 4.2 K. Analysis with an X-ray spectrometer showed that there is no pronounced texture in the alloy samples; therefore, these samples can be considered polycrystalline. The yttrium and holmium crystal structure in the considered temperature range is hexagonal close-packed (hcp). Before the measurements, the samples were annealed in a chamber at 1400 K for 2 h.

The experimental samples were cut on a DK7732 wire-cutting erosion machine. The total number of studied materials with different compositions was 18: two pure REMs (yttrium and holmium) and 16 alloys with different concentrations. Note that the thermal diffusivities of only eight types of materials were studied in the previous work [8]. In this study, we consider many more materials and characteristics.

EXPERIMENTAL METHODS

The thermophysical characteristics (thermal diffusivity and relative specific heat) were studied by the temperature-wave method [14−16]. This method has been known for a long time (for more than 150 years) and has a number of undoubted advantages for the measurement of thermal characteristics in the high-temperature range in comparison with all of the known methods [17, 18]. The main advantages of this method are the ability to use small samples and to take measurements with low thermal loss, as well as the internal possibility of control [19−22].

The temperature waves in a planar sample were formed by the effect of the amplitude-modulated radiation from an LGN-701 laser (radiation wavelength 10.6 μm, power 60 W) on the first surface of the planar sample. The modulation was performed mechanically with the use of a rotating disk with holes. The disk was set in motion by an AC electronic motor with a Delta ASD-A0121LA servo drive controlled by a computer program [23]. The ability to control the modulation frequency (i.e., the temperature wave frequency) was thus provided. The sample was in a vacuum chamber in a cell, in which a special spacer made of sheet molybdenum was used to prevent sample−cell interaction. The sample temperature was varied with an electric resistance furnace. The current strength was controlled by the heater with a computer.

The temperature of the second flat sample surface was transformed into an electrical signal with two sensors (thermocouple and photoelectric) [24]. A thermocouple sensor (VR5/20) made of a tungsten−rhenium wire 50 μm in diameter was welded without junction [25] near the central point of the second sample surface. The photodetector-sighting region also included this central point, which was located on the optical axis of the system set by the laser beam. Both sensors were used to measure temperature oscillations on the second sample surface. The average sample temperature was estimated from the constant component of the thermocouple voltage.

The sensor signals passed through linear normalizing chains [26], were fed to an analog-to-digital converter, and then were processed in the computer. Data on the phase of the modulated laser radiation acting on the sample were also sent to the computer. The amplitude and phase shift of temperature oscillations on the second sample surface (with respect to the laser radiation oscillation phase) were estimated according to the quasi-optimal procedure via Fourier transform [27].

The thicknesses of the plane-parallel samples in our measurements were chosen from the range of 0.8−1.8 mm. The temperature wave frequencies were changed from 6 to 16 Hz. Estimation of the measurement errors showed that the uncertainties of measuring the thermal diffusivity and relative specific heat are 2% and 5%, respectively (mean-square values).

The resistance was measured according to Ohm's law on DC in a setup with the four-probe (four-wire) method [28]. The measurement error for resistance was estimated to be 2.7% (the mean-square value).

The techniques of the experimental determination of the thermophysical properties and electrical resistance were certified by the All-Russia Research Institute of Metrological Service [29, 30].

EXPERIMENTAL RESULTS

In the considered temperature range, the thermal diffusivity of the studied materials obeys nearly linear laws. A monotonic increase in thermal diffusivity is observed upon heating. The thermal diffusivity value numerically depends on the component concentrations. The thermal diffusivities of pure Y and Ho were studied previously in [6, 7]. Figure 1 shows the measurement results. In particular, the literature data and the data from this study are presented in Fig. 1a for Y and in Fig. 1b for Ho. Note the satisfactory agreement between these data. Figure 1c gives data on the polytherms of the thermal diffusivity of some alloys. It can be seen that, despite the known concepts, these polytherms are nonparallel. Some of them intersect.

The specific heats of the studied materials in the considered temperature range also increase according to nearly linear laws. The results of relative measurements were bound to absolute values based on the reference data on specific heats of pure Y and Ho [6] and the mass concentrations of the components in alloys. The specific heat polytherms for different analyzed concentrations do not intersect.

The alloy resistances are described by nonlinear functions with negative curvature, i.e., the rate of increase in the electrical resistance decreases with an increase in temperature. Figures 2a and 2b show the data for Y and Ho; one can see that the results are in satisfactory agreement. Figure 2c gives the results of the study of the alloys. These polytherms (as the ther-
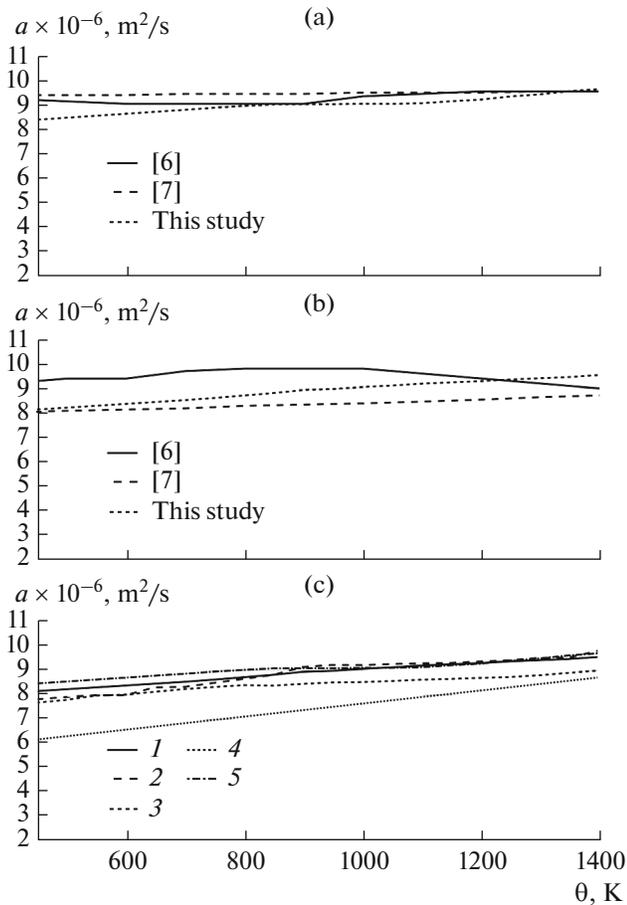
Fig. 1. Thermal diffusivities of pure (a) Y and (b) Ho and (c) of the Y–Ho alloy system: (*1*) 0, (*2*) 30, (*3*) 50, (*4*) 70, and (*5*) 100 wt % of Y.

Fig. 2. Electrical resistivity of (a) Y, (b) Ho, and (c) Y–Ho alloys: (*1*) 0, (*2*) 30, (*3*) 50, (*4*) 70, and (*5*) 100 wt % of Y.

mal diffusivity polytherms) are nonparallel, and some of them intersect.

On the whole, one can see that the studied thermophysical properties qualitatively change (as the corresponding properties of pure REMs); however, they exhibit a quantitative difference, which depends on temperature. Let us consider the reasons for such discrepancies.

DISCUSSION

The studied temperatures exceed significantly the Debye temperatures of the alloy components (the highest is 220 K) [6]. Therefore, the heat capacity of the alloys mainly consists of the temperature-independent harmonic-phonon contribution and the contribution from collective electrons [32]. The experimental results showed that the excess of the molar-lattice heat capacity, which is equal to $3R$ (R is the universal gas constant [32]), is described by the electron contribution. Indeed, the calculation of the molar value of the coefficient of electron heat capacity γ_e performed based on our measurements yields $\gamma_e \approx 4 \times 10^{-3}$ J/(mol K^2). This value does not
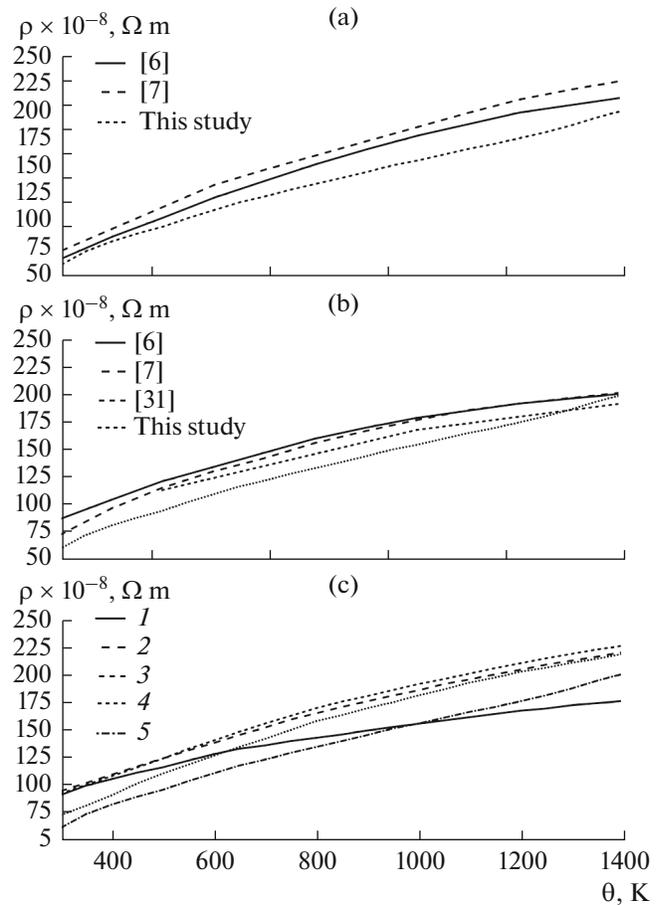
exceed the coefficients of electron heat capacity for yttrium and holmium reported in the literature, e.g., 10×10^{-3} and 8×10^{-3} J/(mol K^2) for Y and Ho, respectively, [33] or 7.9×10^{-3} and 6.0×10^{-3} J/(mol K^2) for Y and Ho, respectively [34]. Thus, the contributions of harmonic phonons and electrons describe the process of energy accumulation by Y–Ho alloys up to 1400 K. Other mechanisms do not manifest themselves at these temperatures.

Let us consider the kinetic properties. First, we should determine the mechanisms of heat transfer that are significant in Y–Ho alloys at high temperatures. To this end, it is convenient to consider the thermal conductivities of the alloys. The temperature dependences of the thermal conductivity $\lambda(T)$ are plotted based on the experimental data on the thermal diffusivities, heat capacities, and densities and calculated in correspondence with the definition of thermal diffusivity [32]. The alloy densities were found from the densities of the pure components [35–37] with allowance for their mass fraction in the alloys.

The corresponding calculation showed that the thermal conductivities of Y–Ho alloys in the considered temperature range monotonically increase with different curvatures. The estimation of the electron components of the thermal conductivity performed based on the Wiedemann–Franz law [32] revealed that the thermal conductivities of the alloys are mainly determined by the electron mechanism. It is responsible for specific features of the behavior of the total thermal conductivity; therefore, the physical reason for the observed behavior of the thermal conductivity is the same as that for the resistance. Thus, we should determine why the resistance of Y–Ho alloys changes in the way shown in Fig. 2.

The character of changes in the properties of Y–Ho alloys upon heating is qualitatively described by the two-band conductivity model (*sd* Mott model) [8]. Moreover, this character is the same on the whole as that for pure REMs [7, 38, 39]. This similarity in appearance makes it possible to develop a procedure to separate the contributions to carrier scattering [40] and, thus, to establish the factors determining specific features of the temperature dependences.

As for pure REMs, we assume that the alloy conductivity is mainly due to light *s* electrons. The transitions of light electrons to the *d* band are most likely to occur during the scattering processes related to interactions between electrons and inhomogeneities, due to which these electrons lose mobility. Within the two-band model, this process leads to an increase in the resistance in comparison with the single-band model [41].

As noted above, the alloy components (yttrium and holmium) have different molar masses and ion magnetic moments. Thus, the process of carrier (electron) scattering in the alloys should be determined by three mechanisms at high temperatures [41, 42]: impurity ρ_{im}, phonon ρ_{ph}, and magnetic ρ_m. Then, in accordance with the Matthiessen rule [41, 42], the total resistance ρ can be presented as the sum

$$\rho = \rho_{im} + \rho_{ph} + \rho_m.$$

In pure metals, the amount of impurities is small and their contribution to the resistance is comparable to the error in measuring ρ; therefore, in this analysis, it can be assumed that $\rho_{im} = 0$ for pure metals. Yttrium is a Pauli paramagnet [13]; therefore, the magnetic resistance is absent therein [4]. In this approach, yttrium is characterized only by phonon scattering, for which $\rho = \rho_{ph}$. In the *sd* model, the phonon resistance can be described by the following formula [38, 40]:

$$\rho_{ph} = Z(\Theta)\frac{\gamma^{1/3}}{\Theta_R^2}\frac{\theta}{M^{4/3}}, \qquad (1)$$

where γ is the density, Θ_R is the kinetic Debye temperature [41], M is the molar mass, and $Z(\Theta)$ are the other parameters, including the temperature-dependent density of states of *d* electrons. In correspondence with (1), the temperature dependence of the phonon

electrical resistance is determined by two factors: an increase in the number of phonons upon heating (parameter Θ) and a decrease in the density of states of *d* electrons upon heating (parameter $Z(\Theta)$). Therefore, formula (1) describes the yttrium resistance.

The characteristics of collective yttrium and holmium electrons are similar [1, 3, 9, 10]. Their crystal structures are identical [3, 6, 13]. These circumstances suggest that the phonon components of the total alloy resistances are also given by formula (1), in which corresponding effective values of parameters γ, Θ_R, and M should be taken into account:

$$M = (1 - x)M_{Ho} + xM_Y,$$

$$\Theta_R = (1 - x)\Theta_{R,Ho} + x\Theta_{R,Y},$$

$$\gamma = (1 - n)\gamma_{Ho} + n\gamma_Y,$$

where x and n are the atomic and mass yttrium concentrations, respectively. Subscripts Ho and Y indicate parameters of the corresponding element.

The calculation from formula (1) showed that the phonon alloy resistances monotonically increase and the growth curve is characterized by negative curvature. The corresponding dependences do not intersect. The phonon resistance of yttrium is maximal, while that of holmium is minimal (as should be expected according to the accepted model [38, 40]).

The resistance of holmium has two significant components (phonon and magnetic): $\rho = \rho_{ph} + \rho_m$. The phonon component was found from formula (1); therefore, we can calculate the magnetic component. The calculations showed that ρ_m has the maximum value (45×10^{-8} Ω m) near room temperature and gradually decreases upon heating almost to zero near the melting temperature.

The magnetic contribution to the electrical resistance is proportional to the magnetic ion concentration [4]. Thus, we have for Y– Ho alloys

$$\rho_{m,(1-x)} = (1 - x)\rho_{m,Ho}. \qquad (2)$$

The calculation from formula (2) showed that the temperature dependences of ρ_m decay upon heating and form a set of nonintersecting curves. The differences between the electrical resistances of the alloys and their phonon and magnetic components are used to calculate the impurity contribution to the scattering.

Thus, all of the main components determining the electrical resistances and, accordingly, the thermal conductivities of Y–Ho alloys are found. The performed analysis show why the temperature dependences of the thermal conductivity and electrical resistance intersect: the magnetic contribution to the scattering. This contribution differs for different alloys and decreases upon heating. In the low-temperature part of the considered range, the ρ_m of holmium is comparable with phonon scattering; thus, the resistance of holmium is higher than that of yttrium. However, this contribution becomes insignificant at high tempera-
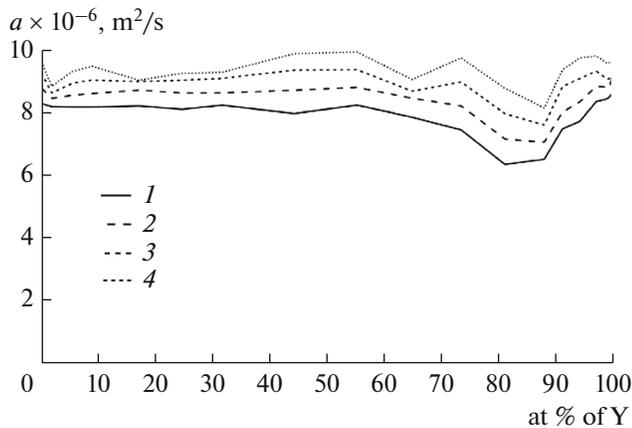
$a \times 10^{-6}$, m²/s

Fig. 3. Concentration dependences of the thermal diffusivities of Y–Ho alloys at different temperatures: (1) 500, (2) 800, (3) 1100, and (4) 1400 K.
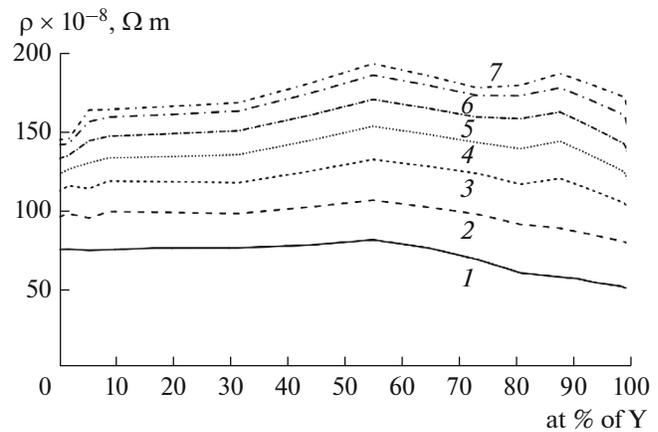
$\rho \times 10^{-8}$, Ω m

Fig. 4. Concentration dependences of the resistance of Y–Ho alloys at different temperatures: (1) 300, (2) 500, (3) 700, (4) 900, (5) 1100, (6) 1300, and (7) 1400 K.

tures, and the yttrium phonon resistance becomes the highest. The latter is related to the fact that the yttrium molar mass is smaller than the effective molar masses of the alloys and that the yttrium ion vibrates with a higher velocity, thus providing a larger scattering value.

Let consider the concentration dependences. Figure 3 shows the thermal diffusivities of the studied alloys. As noted above, the heat transfer in REM alloys at high temperatures is mainly determined by electrons; therefore, it is of interest to consider the concentration dependence of the resistance shown in Fig. 4. For both considered parameters, the concentration dependences are asymmetric, i.e., they do not obey Nordheim's rule [41]. It was shown experimentally that addition of a small amount of yttrium to holmium changes only slightly the scattering properties of the alloys. Conversely, the addition of a small amount of holmium to yttrium significantly amplifies the scattering (the resistance increases). In other words, as noted in [8], "holmium is a more significant impurity for yttrium in comparison with yttrium for holmium."

The reason for this behavior of the considered dependences is clear: magnetic electron scattering. Indeed, the largest contribution to scattering due to disordered magnetic moments is made in holmium. When the latter is diluted with yttrium, this contribution decreases proportionally to the holmium concentration; however, the impurity and phonon contributions increase due to the occurrence of yttrium in the alloy. It was shown experimentally that these effects are almost compensated for; therefore, the resistance of the alloys with low yttrium concentrations barely changes. The addition of holmium to yttrium leads to an increase in scattering due to the magnetic and impurity contributions. Therefore, the electrical resistance increases while moving along the concentration dependence from yttrium to holmium.

The magnetic and phonon contributions to carrier scattering are not symmetrical relative to the compo-

nent concentrations; therefore, Nordheim's rule is not valid for these contributions. It should be noted that the concentration dependence of the magnetic resistance component corresponds to the known concepts [4]; however, the decrease in ρ_m in the paramagnetic phase upon heating cannot be explained with modern magnetic-scattering models.

One might expect Nordheim's rule to be valid for the impurity contribution due to the random positions of ions of different masses in the alloy. Figure 5 presents the corresponding dependence. Indeed, the electrical resistance maximum corresponds to the central region of concentrations (i.e., to the region with the strongest structural disorder).

This result is not trivial. In alloys of transition metals (which includes REMs), the resistance is determined by not only disorder in the system but also the concentration range corresponding to the maximum density of states of heavy d electrons [7, 38–40, 43, 44]. The character of the concentration dependence of the impurity resistance (Fig. 5) indicates that the elec-
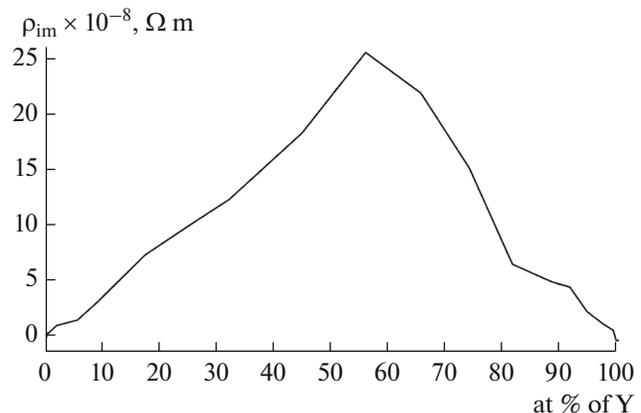
$\rho_{im} \times 10^{-8}$, Ω m

Fig. 5. Concentration dependence of the impurity contribution to the resistance of Y–Ho alloys at 300 K.

tron density of states in the studied alloys barely depends on the component concentrations. It should be noted that the impurity scattering value calculated in this study is close to the results of direct measurements carried out at low temperatures for rare-earth alloys [5, 45].

CONCLUSIONS

The temperature dependences of the thermophysical properties of Y–Ho alloys obey the same laws as the properties of pure REMs. The alloy heat transfer is mainly determined by the electron mechanism. The processes of electron heat and charge transfer in the alloys can be described with the two-band conductivity model (the Mott model). Based on this model, the contributions to carrier scattering from the phonon-, magnetic-, and impurity-scattering mechanisms can be separated. The existing quantitative differences in the properties of the studied alloys are due to unequal influences of different contributions to the conduction electron scattering. Due to this, the concentration dependences of the analyzed properties do not obey Nordheim's rule. This rule is valid for impurity scattering, which indicates the similarity of the electron energy spectrum for all of the studied alloys.

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