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Electric Resistance of Rare-Earth Metals and Their Alloys at High Temperatures: the Role of Magnetic Scattering

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Abstract—Results of an experimental study of the specific resistivity of rare-earth metals and their alloys are analyzed. A procedure to distinguish the contributions into the total resistance is proposed. The magnetic component of electric resistance in the paramagnetic phase was found to decrease to zero upon heating. Modern models of transport phenomena do not describe this phenomenon.

Keywords: rare-earth metals, alloys, specific resistance, magnetic component of resistance **DOI:** 10.1134/S1063783420100145

1. INTRODUCTION

Condensed phases of scandium, yttrium and lanthanides are called rare-earth metals (REMs) [1]. Their chemical properties are similar due to the same energy structure of collective electrons. Nevertheless, all REMs are divided into two groups related to yttrium and cerium. Metals within each of these groups possess especially close properties. Metals of the yttrium groups (i.e., yttrium and heavy REMs from gadolinium to lutetium excluding ytterbium) are considered in this paper.

Information on the specific resistance presented here was obtained by the four-probe method using direct current. The typical measurement principle is described, for example, in [2].

At high temperatures (above the Debye temperature) polytherms of electric resistance of pure REMs [3] and their alloys [4] are similar. The rate of resistance growth becomes smaller as the temperature grows, and the absolute value of the resistivity is relatively high, larger than the typical values of this parameter of other transition metals [3]. An important feature of resistivity polytherms is that they do not form a group of equidistant dependences but rather form a system of crossing curves. This peculiarity is met both for pure REMs and for the investigated alloys.

A question arises, which scattering mechanisms can be significant in the high-temperature resistance of REMs and their alloys? As mentioned above, structure of energy levels of collective electrons in these materials are similar [5-10]. Hence the differences between the resistivity values are related mainly to the properties of the ions [11-13]. Ion masses smoothly increase twofold when REMs in the series from yttrium to lutetium are considered. This should lead to a decrease in the resistance of REM caused by electron scattering on lattice oscillations as we consider heavier metals [11–13]. This is true near the highest temperatures close to the melting point [3, 14, 15]. At intermediate temperatures, however, gadolinium has the largest resistivity among all REMs [3]. Therefore, another (magnetic) mechanism plays an important role in electron scattering in addition to the phonon mechanism in this temperature range [11].

Magnetic properties of REM ions are very different. They are defined by 4*f* electrons, which form atom-type energy levels in the considered phase and do not form energy bands [16, 17]. Magnetic moments of ions in Pauli paramagnetics (yttrium and lutetium) are zero. Any of the other REM ions possesses a magnetic moment consisting of the spin and orbital contributions.

Holmium ion has the largest orbital angular momentum and total quantum numbers [16, 17]. Apparently, holmium should exhibit the strongest contribution of magnetic scattering into resistance. However, this is not true. Furthermore, the temperature of the paramagnetic phase transition of holmium is smaller two times than that of gadolinium (the latter has the highest paramagnetic transition temperature among REMs) [3]. The gadolinium ion have zero angular momentum, but its spin momentum is the largest [16, 17]. Thus, it is the spin magnetism of ions which provides the high resistivity of metallic gadolinium and the higher transition temperature to the paramagnetic state. This statement agrees with the indirect exchange model, in which the spin of the ions is responsible for the magnetic interaction between the REM ions [6, 7, 16, 17].

In addition to the abovementioned phonon and magnetic terms, impurity scattering is also present in REMs and alloys. There are no other significant scattering mechanisms in the considered temperature range [11–13]. According to the Matthiessen's rule [11], the total specific resistance ρ can be written as

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$$\rho = \rho_L + \rho_M + \rho_r, \tag{1}$$

where ρ_L , ρ_M , and ρ_r stand for the phonon, magnetic, and impurity terms. To explain the reasons for the observed behavior of ρ of REMs and alloys, it is necessary to estimate the fractions of these terms in the total resistance. In other words, it is necessary to distinguish between different contributions in ρ .

2. FORMULATION OF THE PROBLEM

2.1. Methods of Separation of the Components of the Resistivity

The separation procedure influences the result and, hence, the physical conclusions. Three methods of distinguishing the terms comprising ρ are used today.

The first approach relies on the assumption that the phonon resistivity of all metals (including transition metals) obeys the Bloch law [11]. According to this rule, ρ_L at high temperatures is proportional to the temperature Θ . Thus, a straight line plotted from the coordinate origin and parallel to the high-temperature segment of the polytherm $\rho(\Theta)$ is ascribed to the high-temperature contribution of phonons. The impurity component ρ_r is considered temperature-independent, and therefore its value was determined from the ratio of the resistivity values of the sample at room temperature and at liquid helium temperature (4.2 K), since all other terms in ρ become negligible at helium temperature [1], ρ_M was calculated then using Eq. (1).

This approach has been widely used for transition metals (see [3, 6, 16, 18]). Results obtained thus are obvious. In REMs the value of ρ_M is much larger than ρ_L . Allowing for the fact that the polytherms $\rho(\Theta)$ are nonlinear and have negative curvature, the value of ρ_L depends on the choice of the temperature range in which the segment of the dependence $\rho(\Theta)$ is considered linear. The larger is the chosen temperature, the smaller is $\rho_L(\Theta)$. The magnetic term is revealed in all REMs. In particular, in nonmagnetic yttrium it is several times larger than the phonon resistance, and is related to a kind of electron–electron scattering [3]. The values of ρ_M is REMs remain quite noticeable until the melting points.

There are theoretical models allowing us to consider the temperature dependence of ρ_M in the paramagnetic phase, in the high-temperature region. Usually they state that ρ_M tends to a constant values (i.e., it is nonzero) [6, 16, 19–21] or increases limitless proportionally to the temperature [22]. Taking into account that the considered approach of distinguishing the terms and the models of magnetic scattering are not perfect, there is no contradiction between the obtained experimental results on the contributions separation and the theoretical results. However, this is the only positive feature of the discussed approach.

The use of the Bloch model for describing the phonon resistance in transition metals is a fundamental drawback of the considered separation method. The Bloch model is based on the single-band model of electron conductivity, and in its standard form [11] it cannot be applied to describe the properties of transition metals having complex electronic structures [11, 12]. Strictly speaking, the single-band model is developed for alkaline metals and does not allow for specific charge transport processes in multi-band systems [23]. For this reason large contributions into the resistance appear in analysis of the results, which are ascribed to the magnetic scattering in the single-band model and in the considered term separation manner. These contributions appear not only in yttrium, but in REMs which are Pauli paramagnetics. It disagrees with existing fundamental results of the transport phenomena theory in solids [11-13].

Results of analysis of the resistance obtained within the first model also do not allow understanding some other peculiarities of the polytherms ρ of REMs and alloys, which will be discussed below (section 3) when the processing of the experimental data will be described. Thus, the first approach of separation of terms in the specific resistance of REMs (and, generally, of transition metals) should be deemed erroneous. The conclusions obtained using this model are erroneous, too.

The second approach for separation the components of ρ is based on the idea that the resistivity of Pauli paramagnetics contains only the impurity and phonon terms. In particular, yttrium and lutetium are Pauli paramagnetic among REMs considered here. Hence, magnetic contribution into the resistivity can be calculated by subtracting ρ of yttrium or lutetium from ρ of magnetic REMs [24, 25]. This approach seems correct from the physical point of view. The magnetic resistivity component obtained this way decreases as the temperature grows [24]. The disadvantage of this method is the p of yttrium at high temperatures is larger than for other REMs, and the difference between the resistivities becomes negative (i.e., it tends to a negative constant as the temperature increases). If ρ of lutetium is subtracted, the difference tends to a positive constant. In other words, this method of distinguishing the components is ambiguous despite its physical clarity.

The third scheme is close to the second in its physical concept and basic results, but it uses renormalization of the resistivities of REMs to avoid the ambiguity. Note that the second and third separation methods can be applied only to materials having similar energy levels of outer electrons in atoms and, hence, collective electrons in the condensed phase. Only REMs suit this requirement among the elements of the periodic table.

The idea of the separating the resistivity components of pure REMs is described, for example, in [26]. It is based on the Mott's model of two-band conductivity [27]; it is the simplest model developed to describe kinetic phenomena in transition metals. Note that the real structure of energy levels of collective electrons in REMs is more complicated than in the described model, but this simplified approach allows concerning main problems in analytical form, which is useful. The model deals with two energy bands of s and d electrons. The first s-band has a relatively low density of electron states $N_{s}(W)$ which depends insignificantly on the energy W. As a result, s electrons have a small effective mass [13]. The second *d*-band is narrow, the electron states density $N_d(W)$ is higher than in the s-band, and it depends stronger on the electrons' energy $N_s(W)$. Therefore, effective mass of d electrons is bigger than that of *s* electrons.

The presence of two energy bands allows rewriting the total conductivity of a material $\sigma = 1/\rho$ as a sum of conductivities of each band:

$$\sigma = 1/\rho = \sigma_s + \sigma_d, \tag{2}$$

where σ_s and σ_d are the conductivities due to the motion of *s* and *d* electrons, respectively. This relation can be simplified at the cost of quantitative accuracy, if we suppose that $\sigma_s \gg \sigma_d$ due to the difference in the effective masses [12]. Then

$$\sigma = 1/\rho \approx \sigma_s = 1/\rho_s, \tag{3}$$

where ρ_s is the resistivity of the conductor provided by *s* electrons. The value of the specific resistance (and hence conductivity) is limited by scattering of charge carriers [13]. Whatever is the nature of scattering, it can either leave an electron in the *s*-bands (*ss* transition, contribution ρ_{ss}) or move it to the *d* band (*sd* transition, contribution ρ_{sd}). Hence,

$$\rho \approx \rho_s = \rho_{ss} + \rho_{sd}. \tag{4}$$

Transition probabilities depend, in particular, on the densities of the electron states in the vicinity of the chemical potential η (i.e., at $W = \eta$) [13]. According to Ziman's estimate [11],

$$\rho_{sd} \approx \rho_{ss} N_d(\eta) / N_s(\eta). \tag{5}$$

Thus, we have [25]

$$\rho \approx \rho_{ss} + \rho_{sd} \approx \rho_{ss} + \rho_{ss} N_d(\eta) / N_s(\eta)$$

= $\rho_{ss} [1 + N_d(\eta) / N_s(\eta)],$ (6)

where ρ_{ss} is the resistivity calculated for the *s*-band only. If expression (6) is used for description of the

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phonon component ρ_L , then ρ_{ss} should be taken, for instance, from the Bloch equation [12]:

$$\rho_{ss}(\theta) = \frac{(3\pi^2)^{1/3}\pi^3\hbar^3}{4e^2k} \frac{N^{1/3}}{n_a^{2/3}\theta_R^2 m_A V^{1/3}} \theta, \tag{7}$$

where n_a is the number of conductivity electrons per atom, m_A is the mass of the ion, N is the number of ions in volume V, θ_R is the kinetic Debye temperature for electric resistance, k is the Boltzmann constant, e is the elementary charge, and Θ is the temperature.

Based on Eqs. (6) and (7), we can write

$$\rho_L = Z(\theta) \frac{\gamma^{1/3}}{\theta_R^2} \frac{\theta}{M^{4/3}},\tag{8}$$

where γ is the density and M is the molar mass of the conductor, and $Z(\Theta)$ contains other parameters including the temperature-dependent density of states of d electrons. The function $Z(\Theta)$ includes parameters which are the same for all considered REMs. The values of γ , M, and Θ_R for pure REMs are tabulated in [3].

The function $Z(\Theta)$ can be calculated knowing the phonon resistivity of yttrium. Then phonon resistivity of lutetium can be found using the table values of density, molar mass, and kinetic Debye temperature. Resistivity obtained this way coincides (within the measurement error) with the experimental value [26]. This proves that the function $Z(\Theta)$ is universal for all analyzed REMs. Thus, phonon resistivity ρ_L can be calculated for every of the considered REMs provided that the function $Z(\Theta)$ is known.

Impurity resistance is evaluated, as usual, from the ratio of resistivities at room temperature and at 4.2 K. Finally, Eq. (1) yields the magnetic contribution for each metal.

In cases of REM alloys (solid solutions), ρ_L is calculated by the same Eq. (8). The problem of finding the parameters such as γ , M, and Θ_R arises since this information lacks in the literature. To a first approximation, these values can be estimated based on the concentration of the components [28]. For Y–Ho (yttrium–holmium) alloys we get

$$M = (1 - x)M_{\rm H_0} + xM_{\rm Y},$$
(9)

$$\Theta_R = (1 - x)\Theta_{R,\text{Ho}} + x\Theta_{RY}, \qquad (10)$$

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

$$x = \frac{n}{n + \frac{(1 - n)M_{\rm Y}}{M_{\rm Ho}}},$$
(11)

where x is the yttrium concentration (atom fraction), n is the mass fraction of yttrium, and indices as γ , M, and Θ_R denote the corresponding metal. Thus, Eq. (8) can be used to calculate the phonon electric resistivity not only of pure metals but also of their alloys. The magnetic component of the specific resistance of alloys can be found based on the value of ρ_M of the magnetic metal in the alloy (holmium in the considered example). The theory shows that the contribution of this term into the resistivity is proportional to the concentration of magnetic ions [9]. Thus, we can write for the Y–Ho system:

$$\rho_{M,(1-x)} = (1-x)\rho_{M,\text{Ho}}.$$
 (12)

Further, the value of the impurity resistance of the alloys may be found knowing ρ_L and ρ_M from Eq. (1).

3. RESULTS AND DISCUSSION

Now direct out attention to the investigations of resistivity of REMs and Y–Ho alloys, reported in [3, 4, 14, 15, 26, 28]. According to the third scheme of distinguishing the resistivity terms, phonon resistivity values ρ_L of pure REMs were calculated from Eq. (8), impurity resistivities ρ_r were estimated, and then magnetic components ρ_M were found. Obtained dependences had expectedly similar shapes and differed only in absolute values. Figure 1 shows the results of calculations for Gd, which has the largest magnetic resistivity.

The plots show that the phonon term ρ_L increases upon heating and tends to the total value of ρ , while the magnetic term ρ_M tends to zero. This behavior is common for all REMs and their alloys.

Magnetic components of the resistivity of REMs having the largest ρ_M values (these are Gd, Tb, Dy, and Ho) are presented in Fig. 2. Analogous results were obtained for alloys. Thus, the role of magnetic scattering in the analyzed materials is negligible at temperature above 1000 K.

This conclusion can also be made from the dependences of ρ of REMs at two temperatures: room temperature (300 K) and 1500 K, which are shown in Fig. 3.

At room temperature the magnetic component is big. For this reason the resistivities of magnetic metals is larger remarkably than the resistivities of Pauli paramagnetics Y and Lu. At high temperature, however, the situation is different. Evidently, the resistivity of Y becomes the biggest one, and the value of ρ decreases as we move to Lu. This result agrees with the model (8) describing the phonon contribution. The role of the magnetic term becomes unimportant.

A similar conclusion can also be made from analysis of the concentration dependences of ρ for Y–Ho alloys, which are plotted in Fig. 4. At low temperatures ($\Theta = 300$ K) the resistivity of Ho (x = 0) is larger than that of Y. This is due to magnetic scattering. As the concentration x of yttrium grows, ρ remain almost constant until x = 60%. The increase in the yttrium content and decrease in the magnetic resistivity (12) are compensated by the growth of the impurity resis-



Fig. 1. Specific resistance of Gd and its components: (1) ρ , (2) ρ_L , and (3) ρ_M .



Fig. 2. Magnetic term ρ_M in resistivities of (1) Gd, (2) Tb, (3) Dy, and (4) Ho.



Fig. 3. Total resistivity of REMs of the yttrium group at (1) 300 and (2) 1500 K.

tivity, which is due to the increasing structural disorder in the system. Experiment shows that these two trends are balanced, and hence ρ remains almost unchanged.

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Fig. 4. Concentration dependences of the total resistivity of the Y–Ho alloys ((1) 300 K, (2) 1400 K).

The resistivity decreases at x > 60%. This is due to decreases in both magnetic scattering and impurity resistivity. The impurity-caused scattering, according to analysis [28], occurs in the middle of the concentration dependence. Thus, the role of the magnetic scattering is noticeable at 300 K. At 1400 K the situation changes. Holmium has smaller resistance than yttrium. The concentration dependences shows only the effect of structural disorder (Nordheim's rule: the resistivity maximum is near x = 50% [11]). The influence of magnetic scattering is not observed.

The experimentally found crossing of polytherms ρ of pure REMs [3, 14, 15] and their alloys [4, 28] is caused by the fact that as the temperature grows, the value of ρ_M almost disappears, and lattice oscillations become the main scattering factor.

The discussed peculiarities of the concentration and temperature dependences of the specific resistances of REMs and their alloys cannot be explained if ρ_M is assumed constant at high temperatures (like it is suggested in the first approach of distinguishing the components).

Thus, existing experimental data allow proposing that magnetic resistivity in paramagnetic REMs and their alloys contributes significantly into ρ only at intermediate temperatures. At high temperatures this component is negligible.

4. DISCUSSION

The problem arising in analysis of the temperature dependence of ρ_M for REMs and their alloys is the absence of a physical model which could explain the decrease in the magnetic resistivity of paramagnetic metals upon heating. It is worth noting that at low temperatures, when magnetic structures of REMs are ordered, the models also do not describe the temperature dependence $\rho_M(\Theta)$ accurately enough.

This is the best illustrated by analyzing the properties of gadolinium: is has the largest ρ_M , and hence the



Fig. 5. Magnetic term ρ_M in resistivity of Gd in the ferromagnetic and paramagnetic temperature regions.

relative error will be minimal. The term ρ_M will be separated following the third approach, and ρ_L will be calculated using Eq. (8). The literature data for ρ from [3] will be taken for the low-temperature region. The result is shown in Fig. 5.

Evidently, ρ_M is an additional contribution into ρ , providing an increase in the resistivity over the phonon and impurity terms. The maximal value of ρ_M corresponds to the Curie point Θ_C [3]. The dependences of resistivity upon heating are different in the ferromagnetic and paramagnetic regions. Accordingly, the physical reasons responsible for the increase and decrease in ρ_M are also not the same. Approximation of obtained results gives the following relations:

for
$$\Theta < \Theta_{\rm C}$$
: $\rho_M = 14 \times 10^{-11} \Theta^{3/2}, \Omega \, {\rm m},$ (13)

for
$$\Theta > \Theta_{\rm C}$$
:
 $\rho_M = 145 \times 10^{-8} \exp(-2.2 \times 10^{-3} \Theta), \, \Omega \, {\rm m}.$
(14)

Experiment shows that ρ_M grows according to the power law (13). This type of the polytherm agrees generally with the present theoretical concepts on the role of magnetic scattering [6, 16]. The power index 3/2, however, appears only in an exotic case when electrons are scattered on two-dimensional spin waves near domain interfaces [16]. There is still no general solution of this problem, which would estimate ρ_M of a ferromagnetic metal in the slope of the multi-band conductivity model.

At high temperatures $\Theta > \Theta_C$ the magnetic scattering diminishes almost to zero following the exponential law (14). No ab initio models have been developed by now which would help to understand this fact. It was stated above that theoretical calculations predict that ρ_M should either be constant or even increase upon heating in the paramagnetic state. The paramagnetic phase is considered as the state with the maximal magnetic disorder of the system; due to this assumption, ρ_M achieves its maximum at Θ_C in most of the models and remains constant at further heating. Thus, existing models disagree with the experiment.

At the same time, the constant value of ρ_M in the high-temperature region follows from the approximations made. In particular, consider the classical result of estimating ρ_M [6]. The resistance value in the paramagnetic state is defined by the de Gennes function *G*:

$$G = (g_J - 1)^2 J(J + 1), \tag{15}$$

$$\rho_M = \frac{3\pi N m_e^*}{8\hbar e^2 \eta V} A_0^2 G, \qquad (16)$$

where g_J is the Lande factor [6], J is the total quantum number of the REM ion [17], A_0 is the exchange integral, N in the number of ions in volume V, m_e^* is the effective mass of the conduction electron, η is the chemical potential (Fermi level) of the conduction electrons, and e is the elementary charge. All values in Eq. (16), including the exchange integral A_0 , are constants. In reality, however, this parameter of the indirect exchange depends on the distance between the metal ions. The spin density of conduction electrons is known to be an oscillating function depending on the distance in the situation with indirect exchange [6, 17, 29] (similar to oscillating density of the electric charge of the shielded impurity potential [30]). The exchange integral should also depend on the distance between ions and, thus, on the temperature. This fact should make ρ_M temperature-dependent. Detailed calculations allowing for the variability of the exchange integral, unfortunately, have not been made; the processes of multi-band scattering have not been considered also in estimates of ρ_M .

Note also that estimates of ρ_M suffer from differences between the experiment and theoretical concepts not only for REMs but also for other transition metals and alloys [31].

Thus, the problem of adequate description of the magnetic component of electron scattering in transition metal, including rare earths, and their alloys in a wide temperature range remains unsolved. This is a fundamental problem, and its solution is necessary for understanding the physics of magnetism and transport phenomena in transition metals at high temperatures.

CONCLUSIONS

Experimental investigation of the resistivity of REMs and their alloys revealed that the magnetic component of the electric resistance in the paramagnetic sold phase decreases upon heating.

Methods of theoretical description of magnetic scattering known so far contradict the obtained result.

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CONFLICT OF INTEREST

The author declares that he has no conflicts of interest.

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