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Thermal conductivity of REIn₃ compounds

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Abstract

The results of measurements of the thermal conductivity of REIn₃ (RE = Pr, Nd, Dy, Ho, Tm) compounds as a function of the temperature in the interval 4–300 K in the absence and in the presence of an external magnetic field of 8 T are presented. Except for PrIn₃ all the compounds are antiferromagnetic. YIn₃ was also measured as a reference compound. The results were analysed in the paramagnetic phase, where an influence of the crystalline electric field on the thermal conductivity was found. Drastic changes in the thermal conductivity were observed and analysed in the vicinity of the Néel temperature and in the antiferromagnetic phases of the compounds. Below the Néel temperature an additional magnon contribution to the thermal conductivity was separated out.

1. Introduction

The thermal conductivity of elemental rare earth (RE) metals and rare earth intermetallic compounds has been examined for many years [1–3]. Nevertheless, there is lack of systematic investigations into the thermal conductivity of high-purity monocrystalline samples of a simple crystalline structure for which the magnetic and electronic properties are well known. Our previous investigations [3] were devoted to the cubic intermetallics REIn₃ with RE = Tb, Dy, Tm and Lu. The measurements were made in the absence of an external magnetic field and at temperatures in the interval 4–80 K. Therefore, it seemed interesting to widen these investigations: (i) by examining other compounds of the same crystal structure, (ii) by broadening the temperature interval and (iii) by studying the influence of an external magnetic field. Thus, in the present paper we investigate REIn₃ (RE = Pr, Nd, Dy, Ho, Tm) at temperatures up to 300 K as well as some of them at low temperatures but in the presence of an external magnetic field of strength 8 T.

Rare earth materials can find applications in certain electronic devices and knowledge of how their thermal conductivity is influenced by the temperature and the external magnetic field seems necessary. By examining monocrystalline samples we are able to get information on their transport properties which are free of the influence of the crystal grain structure. We believe

that by gaining basic knowledge about thermal transport phenomena and by understanding their mechanisms one should be able to produce materials with the required thermal properties.

All the mentioned compounds REIn₃ crystallize in the face-centred cubic (fcc) structure of the type AuCu₃. They order antiferromagnetically at low temperatures. Their Néel temperatures, T_N , are low and do not exceed 20 K. The highest one is that of DyIn₃ ($T_N = 19.35$ K). The scattering in these materials, which is responsible for their electrical and thermal resistivity, is—besides the impurity potential and phonon scattering of the conduction (s-)electrons—isotropic s–f and aspherical (multipole) Coulomb scattering. The interactions responsible for this scattering contribute to the magnetic part of the electrical and thermal resistivity of the materials in question provided this part can be extracted from the total measured resistivity using the Mathiessen rule. The scattering is under the influence of the crystalline electric (CF) field, which splits the $(2J + 1)$ degenerate ground state multiplet of the 4f-shell of the rare earth ions and allows the conduction electrons to gain or to lose an amount of energy equal to the energy of the splitting in the course of scattering in the paramagnetic phase (and that of a CF excitation and/or a spin wave excitation in the ordered phase). The CF parameters and the energies of CF excitations, the magnetic structure and properties, the electronic structure as well as the electrical resistivity and the thermoelectric power of the compounds which we examine in the present paper have already been examined. In particular we know: (i) the crystal field level schemes and energies from neutron scattering [4–6], the Schottky specific heat anomaly and the temperature dependence of the magnetic susceptibility as well as the magnetization isotherms; (ii) the magnetic structures (from the simplest collinear structure in the case of HoIn₃ to the most complicated incommensurate one in the case of NdIn₃); (iii) the electronic structures, the Fermi surface (FS) geometries, the density of states and the cyclotron masses of the conduction electrons at extreme cross-sections of FS, which allow one to estimate the electron–phonon and electron–magnon strength (see the review [7] and papers quoted therein). There are also introductory theoretical considerations on the influence of the crystal field on thermal conductivity [8].

Single crystals were grown by slow cooling of the melt with a stoichiometry of 90 at.% In and 10 at.% RE. The purity of the components was 99.999% and 99.99% for In and RE metals, respectively. The cooling rate was gradually increased from 0.5 K h^{−1} at the beginning of the growing process to 4 K h^{−1} at the end. Rectangular single crystals with edges oriented along fourfold axes with an accuracy better than $\pm 2^\circ$ were used for measurements.

The thermal conductivity was measured using the stationary heat flux method in the temperature range 4–300 K. The experimental set-up and the measurement procedure have been described in detail in [9, 10]. The temperature gradient along the sample was in the range 0.1–0.5 K. Particular care was taken to avoid parasitic heat transfer between the sample and its environment. The measurement error was below 2% and the surplus error estimated from the scatter in the measurement points did not exceed 0.3%.

The measurements of the thermal conductivity in the external magnetic field and in its absence were made in a cryostat constructed for the cooperation with PPMS [11]. The external magnetic field was parallel to the temperature gradient.

The results of the measurements of the electrical resistivity of the REX₃ single crystals can be found in [12, 13]. In the present paper the Wiedemann–Franz (WF) law is used for estimating that part of the total thermal conductivity which is carried by the conduction electrons. This means that the electronic thermal conductivity as a function of temperature is calculated by using the formula $\lambda_e(T) = (L_0 \times T)/\rho(T)$, where $L_0 = 2.45 \times 10^{-8}$ W Ω K^{−2} and $\rho(T)$ denotes the temperature dependence of the electrical resistivity. The obtained values of $\lambda_e(T)$ indicate that heat transport in these compounds is mainly electronic. The contribution of other heat carriers is only a few per cent. The values of the electronic thermal conductivity obtained

in this way are valid only when the relaxation time of the thermal transport and that of the electrical transport are the same, i.e. when the scattering can be assumed to be elastic. This occurs only at sufficiently low temperatures, when the conduction electrons are scattered on impurities and physical defects or at high temperatures exceeding the Debye (θ_D) temperature $T/\theta_D \geq 1$. At intermediate temperatures the discrepancies can be considerable and lead to erroneous estimations of the contribution of the conduction electrons to heat transport. In order to explain the latter statements we write the expression for the total thermal resistivity

$$1/\lambda_e = W_e = W_{ep} + W_{ei} = \frac{3}{C_e(T)v_F^2}[\tau_{ep(\lambda)}^{-1}(T) + \tau_{ei}^{-1}], \quad (1)$$

$$\rho = \rho_{ep} + \rho_{ei} = \frac{m}{n_e e^2}[\tau_{ep(\sigma)}^{-1}(T) + \tau_{ei}^{-1}] \quad (2)$$

where the indices ei denote the temperature-independent electron–impurity (defect) scattering and ep(λ) and ep(σ) stand for the electron–phonon scattering in the case of the thermal and electrical currents, respectively. We use the standard notation σ for the electron conductivity and assume that the electron–impurity (defect) scattering is the same for both the currents. C_e , v_F denote the electronic specific heat and the electron velocity at FS. The electron mass, charge and density are denoted respectively by m , e , n_e . When writing the above expressions (1) one assumes the validity of the Mathiessen rule for the scattering of both the electrical and heat currents, the Klemens laws for the thermal resistivity components and the Drude formulae for the electrical resistivity components. The relaxation times of (1) are dependent on the temperature and the respective dependences follow $\tau_{ep(\lambda)}^{-1}(T) \propto T^3$ for $T \ll \theta_D$, $\tau_{ep(\lambda)}^{-1}(T) \propto T$ —for $T \geq \theta_D$, $\tau_{ep(\sigma)}^{-1}(T) \propto T^5$ for $T \ll \theta_D$ and $\tau_{ep(\sigma)}^{-1}(T) \propto T$ for $T \geq \theta_D$ [14, 15]. It is seen that at a temperature equal to approximately θ_D the temperature dependences of the relaxation time describing the electrical and thermal transport are the same and that they differ considerably at low temperatures. By comparing the change in the conduction electrons during the scattering to the Fermi energy for a given scattering source one can classify the scattering process as inelastic, elastic or quasielastic. An analysis of the temperature dependence of the Lorenz number $L(T)$ can also be used for such a classification [2].

There are other types of electron scattering, which are not included in (1) which are in general inelastic. For the compounds being considered it is, first of all, the magnetic s–f scattering between the conduction electron spin and the spin of the f-shell of the rare earth ions which is influenced by the magnetic order (electron–magnon scattering in their antiferromagnetic state) and the crystal field (see, e.g., the review [16]). There is also the possible influence of quadrupolar (or multipolar) Coulomb (aspherical) potential scattering [16]. The heat current carried by magnons requires separate consideration [2].

2. Results and discussion

Figure 1(a) shows the temperature dependence of the thermal conductivity, $\lambda(T)$, for six samples of REIn₃ on a semilogarithmic scale. At low temperatures, $T \leq 0.1\theta_D$, the magnitude of the thermal conductivity of TmIn₃ is about 1.5 W K⁻¹ m⁻¹ while it is as high as about 70 W K⁻¹ m⁻¹ for YIn₃ (a Pauli paramagnet) which serves as a reference for estimating the background phonon scattering. At temperatures of the order of the Debye temperature, $T \approx \theta_D$, the lowest values of $\lambda(T)$ are about 20 W K⁻¹ m⁻¹ for TmIn₃ and NdIn₃ and the highest one, about 100 W K⁻¹ m⁻¹, is for YIn₃. At intermediate temperatures there is a distinct maximum of the thermal conductivity for NdIn₃, PrIn₃ and YIn₃. In figure 1(b) we present the experimental data for the electrical resistivity, $\rho(T)$, of the examined compounds. The values of the resistivity are in the interval 0.3–8 $\mu\Omega$ cm for all the samples except for NdIn₃.

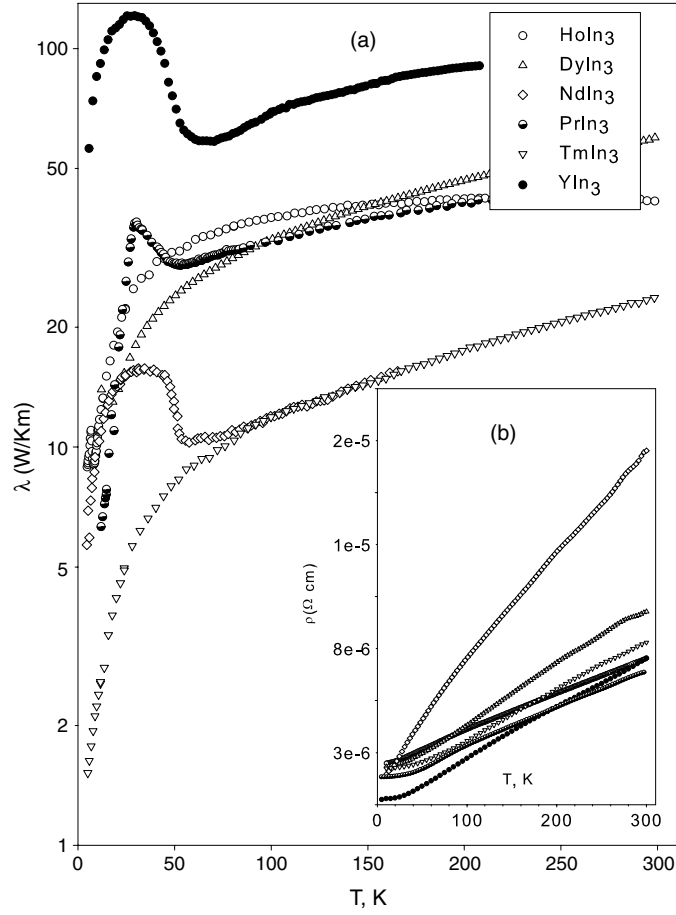


Figure 1. (a) The temperature dependence of the thermal conductivity of REIn₃ and the reference compound YIn₃.

As we mentioned, the measurements of the electrical resistivity were made in order to estimate the electronic thermal conductivity.

2.1. The contribution of spin disorder to the thermal conductivity

In order to extract the spin-disorder thermal resistivity, i.e. the part of the thermal resistivity following from the s-f scattering in the paramagnetic phase, from the total measured thermal resistivity we shall use the method proposed by Bauer *et al* [17]. For this purpose we first write down the expression for the thermal resistivity of the background non-magnetic metal (upper index NM, YIn₃ in our case), which is [17]

$$1/\lambda^{\text{NM}} = W_e^{\text{NM}} = W_{e,i}^{\text{NM}} + W_{e,p}^{\text{NM}}. \quad (3)$$

$W_{e,i}^{\text{NM}}$ and $W_{e,p}^{\text{NM}}$ can be easily identified with terms of (1). An additional term following from the magnetic scattering $W_{e,\text{mag}}$ contributes to the thermal resistivity of REX₃ compounds with an incomplete 4f-shell of R-ions (upper index M)

$$1/\lambda^{\text{M}} = W_e^{\text{M}} = W_{e,i}^{\text{M}} + W_{e,p}^{\text{M}} + W_{e,\text{mag}}, \quad (4)$$

where the lower indices have the same meaning as in (1). Consider now the difference

$$\Delta W = W_e^M - W_e^{NM} = (W_{e,i}^M - W_{e,i}^{NM}) + (W_{e,p}^M - W_{e,p}^{NM}) + W_{e,mag} \quad (5)$$

and note that for different samples, which we discuss, the difference in the first bracket does not vanish but we can assume in the first approximation that the value in the second bracket is equal to zero for samples of the same structure. Therefore, we have

$$\Delta W = (W_{e,i}^M - W_{e,i}^{NM}) + W_{e,mag}. \quad (6)$$

The relaxation time for the impurity scattering is independent of the temperature and the electronic specific heat at sufficiently low temperatures is linear in T (compare (1)). Thus, we can write down

$$\Delta W = (A^M - A^{NM})/T + W_{e,mag}. \quad (7)$$

The constants in (7) are $A = \rho_{ei}/L_0$ with appropriate upper indices, so their values are to be determined from the magnitudes of the residual resistivity of the non-magnetic (YIn₃) sample and the magnetic RIn₃ sample, respectively. The Néel temperatures T_N of the crystals under considerations are very low; thus we can assume the validity of the above equation in the paramagnetic phase of our crystals. When one assumes, furthermore, that the magnetic part of the electrical resistivity arises from the s-f scattering, then in the paramagnetic phase at temperatures higher than the total crystal field splitting the resistivity should be proportional to the de Gennes factor $(g - 1)J(J + 1)$ and then $W_{e,mag} = C(g - 1)J(J + 1)/T$, where C is a constant. Thus, after [17], we can write:

$$\Delta W = (A^M - A^{NM})/T + C(g - 1)^2 J(J + 1)/T. \quad (8)$$

The above equation indicates that the experimental relation between the values of ΔW and $1/T$ should be linear in $1/T$ for our compounds above T_N provided that it is the magnetic s-f scattering which is responsible for the additional thermal resistivity and that the influence of the crystal field on the scattering can be neglected. The dependence ΔW on T^{-1} for REIn₃ (RE = TmIn₃, PrIn₃, NdIn₃, HoIn₃, DyIn₃) following from results of our measurements in the paramagnetic phase is shown in figure 2. The values of ΔW were computed as the difference, $1/\lambda^M - 1/\lambda^{NM}$, between the thermal resistivity of a given RE compound and that of YIn₃. The highest values of ΔW are seen for TmIn₃ and NdIn₃. Equation (8) describes well these experimental data in the temperature intervals 300–180 K for TmIn₃ and 300–75 K for NdIn₃. The magnitudes of ΔW are considerably lower for the remaining three compounds, namely PrIn₃, HoIn₃ and DyIn₃. As in [17], we attribute the departure from the linear dependence (8) at lower temperatures to an influence of the crystal field which for the compounds under consideration is examined, for example in [18–20]. Notice, however, that other magnetic scattering than the assumed s-f one in (8) can also yield a departure from the linear dependence. As seen from figure 3, L/L_0 does not exceed 1 in the whole examined temperature interval, which means that the magnetic scattering is inelastic.

2.2. Investigations of thermal conductivity at low temperatures (the vicinity of $T_N > T \cong T_N$) in the presence and absence of an external magnetic field

Of the examined compounds only PrIn₃ does not manifest a magnetic order down to the lowest temperatures. The antiferromagnetic state in TmIn₃, NdIn₃, HoIn₃ and DyIn₃ forms below T_N , being respectively equal to 1.4, 5.9, 7.9 and 19.3 K. The formation of a magnetic order below T_N leads to lowering of the thermal resistivity [3] and a change in the temperature dependence $\lambda(T)$ due to less intensive scattering. The fact that magnons become heat carriers below T_N is the second factor which causes lowering of the thermal resistivity. Figure 4(a) is a

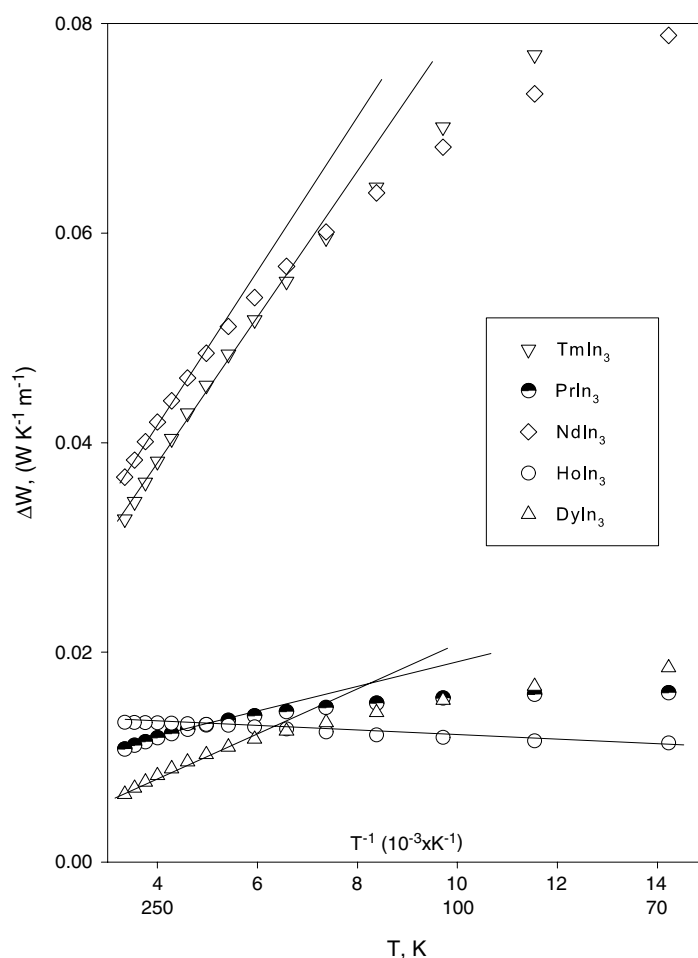


Figure 2. The difference $\Delta W = 1/\lambda^M - 1/\lambda^{NM}$ versus $1/T$. λ^M is the thermal conductivity of $REIn_3$ and λ^{NM} that of the reference compound YIn_3 . The corresponding values of the temperature are marked below those of $1/T$.

double logarithmic plot of the temperature dependence of the thermal conductivity of $DyIn_3$ in the absence of an external magnetic field and in the presence of a field of the strength 8 T. The change in the behaviour of $\lambda(T)_{H=0T}$ is seen at a temperature of 17 K, i.e. at a temperature 2 K lower than T_N determined from the analysis of the magnetic susceptibility. The illustrated change is characteristic for magnetic phase transitions in ferromagnets and antiferromagnets [1–3]. Above T_N , in the temperature interval $50\text{ K} > T > T_N$, the thermal conductivity $\lambda(T)_{H=0T}$ increases as $T^{0.6}$ and below T_N its decrease is governed by the law $T^{-0.39}$. In the external magnetic field of strength 8 T, the thermal conductivity coefficient of $DyIn_3$ is lower than in the field-free case in the vicinity of T_N . The external magnetic field lowers (by about $1.8\text{ W K}^{-1}\text{ m}^{-1}$) the magnitude of the thermal conductivity considerably and shifts the minimum about 0.8 K in the direction of lower temperatures. The power laws which govern the temperature dependence in the presence of a magnetic field are $T^{0.75}$ above T_N and $T^{-0.43}$ below T_N .

Figure 4(b) (double logarithmic scale) illustrates the dependence of the Lorenz function $L = \lambda\rho/T$ on the temperature. The values of L have been calculated by using the measured

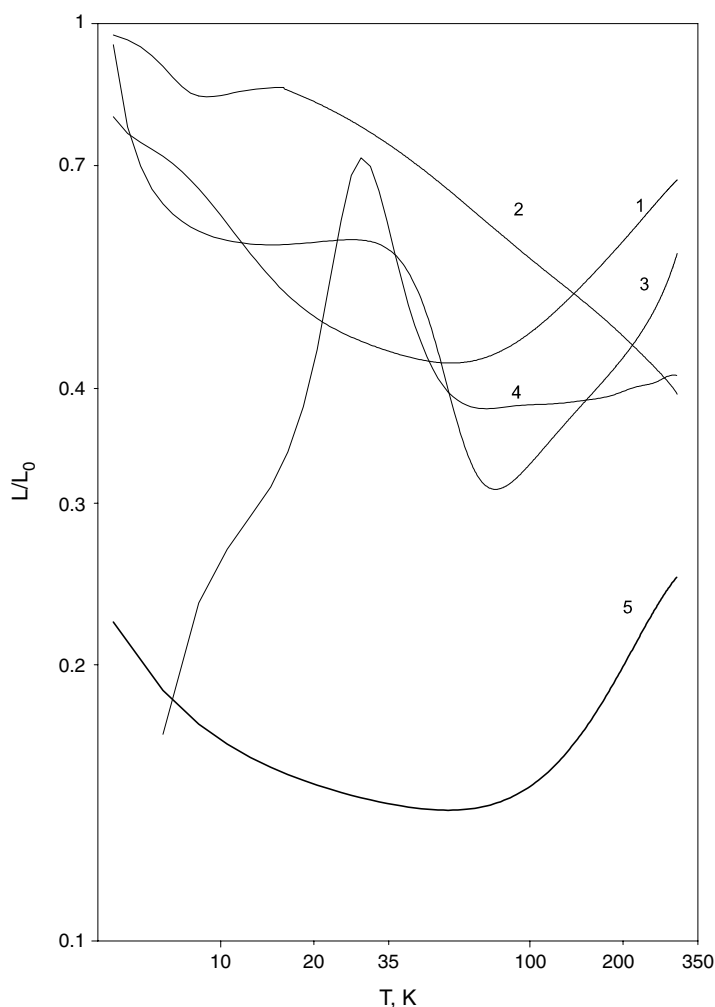


Figure 3. The log–log plot of the Lorenz number L/L_0 (in units of $L_0 = 2.45 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}$) as a function of the temperature for: 1, DyIn₃; 2, HoIn₃; 3, NdIn₃; 4, PrIn₃; 5, TmIn₃.

magnitudes of the total thermal conductivity (λ) and of the total electrical resistivity (ρ) and are represented in units of the Sommerfeld value L_0 of this quantity. It is seen that the values of this function are the same in the vicinity of the transition and below T_N for both the presence and the absence of the field. Differences occur above 30 K. The values of $L/L_0 < 1$ manifest an inelastic scattering contribution to the thermal conductivity; one can expect a contribution to the scattering of the conduction electrons from phonons, magnons and crystal-field excitons [2, 21]. Figure 5(a) shows a double logarithmic plot of the temperature dependence of the thermal conductivity for NdIn₃ ($T_N \approx 6$ K). Unlike DyIn₃, for this compound the minimum of $\lambda(T)_{H=0T}$ is at the temperature of the para–antiferromagnetic phase transition determined from the analysis of the magnetic susceptibility. The power laws which govern this temperature dependence are $T^{0.51}$ above T_N and $T^{-0.58}$ below T_N . The external magnetic field lowers the minimum by about $0.9 \text{ W K}^{-1} \text{ m}^{-1}$ and shifts it about 0.4 K in the direction of lower temperatures. It is seen from figure 5(b) that the values L/L_0 for NdIn₃ are also below 1. The

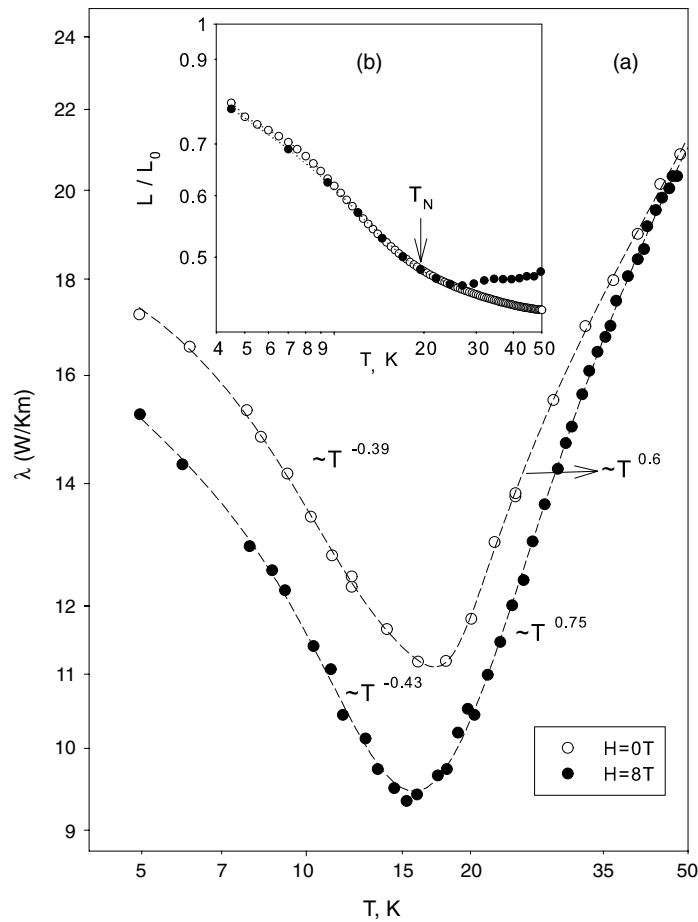


Figure 4. The thermal conductivity (a) and the Lorenz number (b) of DyIn_3 as a function of the temperature in the vicinity of T_N . The interpolation by Chebyshev polynomials in (a) is marked by the dashed lines.

influence of the magnetic field ($\lambda(T)_{H=0\text{T}} > \lambda(T)_{H=8\text{T}}$) on the temperature dependence of L/L_0 is seen at all temperatures except in the close vicinity of the transition temperature.

Similar graphical representations of the thermal conductivity as in the two previous cases are shown in figures 6(a), (b) for HoIn_3 . The behaviour of the dependences $\lambda(T)_{H=0\text{T}}$ and $\lambda(T)_{H=8\text{T}}$ in this case are different, as in DyIn_3 and NdIn_3 . Namely, there are maxima of $\lambda(T)_{H=0\text{T}}$ at about 6.5 K and $\lambda(T)_{H=8\text{T}}$ at about 6.1 K as can be seen in figure 6(a). The difference between the temperature at which the minimum and the maximum occurs is about 1.5 K both in the absence and presence of the magnetic field. The maximum is lowered by the magnetic field about 0.5 K. For HoIn_3 the power laws which describe the behaviour of $\lambda(T)_{H=0\text{T}}$ and $\lambda(T)_{H=8\text{T}}$ are also indicated in figure 6(a); just below T_N these are $T^{-1.37}$ and $T^{-1.19}$, respectively. This means that the decrease is more rapid, as in DyIn_3 and NdIn_3 . The increase above T_N is also steeper since it is governed by $T^{1.05}$ and $T^{0.86}$, respectively. Figure 6(b) is a double logarithmic plot of the dependence of L/L_0 on the temperature for HoIn_3 . A change in this dependence under the influence of the magnetic field is seen not only above the transition temperature but also below it and in its vicinity. The character of the dependences shown is similar to those observed in metals with physical

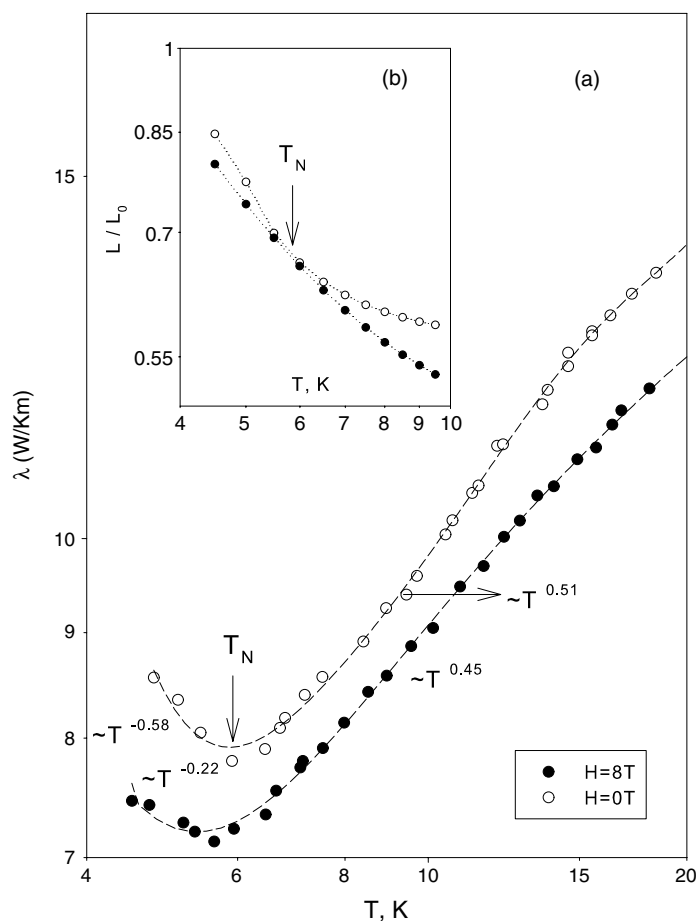


Figure 5. The thermal conductivity (a) and the Lorenz number (b) of NdIn₃ as a function of the temperature in the vicinity of T_N . The interpolation by Chebyshev polynomials in (a) is marked by the dashed lines.

defects and chemical impurities [21]. In metals with such defects and impurities the minimum of L/L_0 results in general from competition between the inelastic electron–phonon scattering and elastic electron-defect and electron-impurity scattering. In rare earth metals the effect can be enhanced by magnetic scattering, which we have briefly described, and perhaps even by the presence of antiferromagnetic domains. The magnetothermal conductivity, defined as $\{[\lambda(T)_{H=0T} - \lambda(T)_{H=8T}]/\lambda(T)_{H=0T}\}$, is presented in a double logarithmic plot in figure 7 as a function of the temperature in the vicinity of the transition temperature. The greatest influence of the magnetic field on the thermal conductivity is seen in the case of HoIn₃ and the weakest temperature dependence of this quantity is for DyIn₃. For NdIn₃ the lowest values of the magnetothermal conductivity are about 8 K.

A further analysis of our experimental data can be made by introducing the notion of magnon thermal conductivity. As we mentioned in the previous section, the magnetic excitations in the antiferromagnetic phase simultaneously transport and dissipate heat. The total effect of the influence of the magnons on the thermal conductivity can be shown by subtracting the thermal conductivity measured in the absence of the magnetic field and in the presence of a field high enough to destroy the antiferromagnetic ordering ($H_{\max} > k_0T/\mu\gamma$;

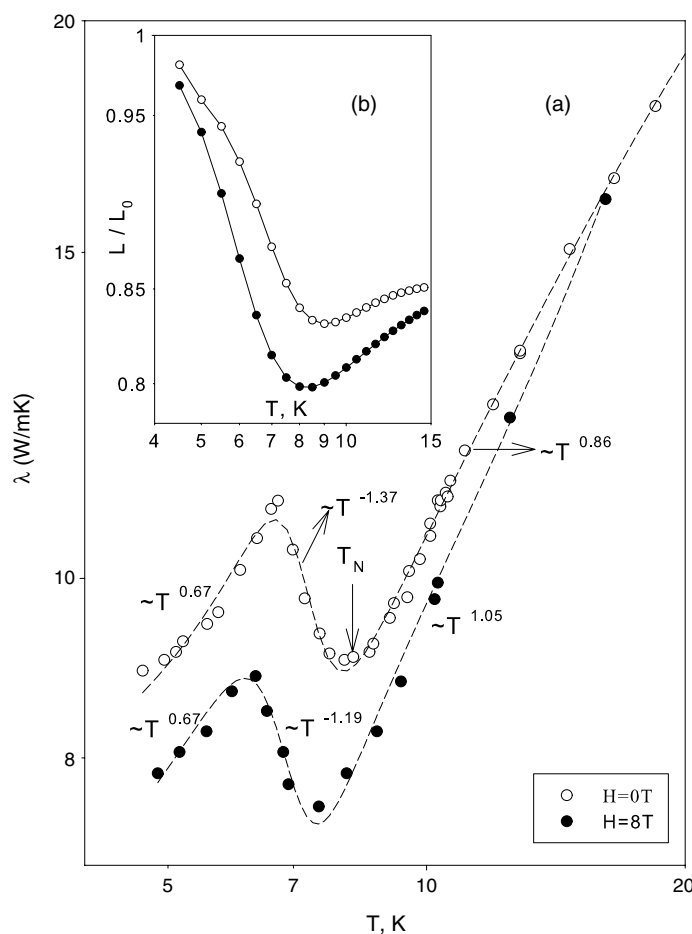


Figure 6. The thermal conductivity (a) and the Lorenz number (b) of HoIn_3 as a function of the temperature in the vicinity of T_N . The interpolation by Chebyshev polynomials in (a) is marked by the dashed lines.

k_0 is Boltzmann's constant and μ and γ the Bohr magneton and gyromagnetic constant, respectively). Separation of the magnon component can also be done by subtracting the thermal conductivity of an *isomorphous* non-magnetic compound or by extrapolation of the thermal conductivity of the paramagnetic state to the ordered one [22, 23].

In order to estimate the magnon thermal conductivity we used the extrapolation method. We applied it to the thermal conductivity in the presence of a magnetic field and in the field-free case and represent the results in figure 8. A rapid increase in the magnon thermal conductivity with lowering of the temperature is observed just below the Néel temperature for all samples. When the number of magnons is decreased with further lowering of the temperature the scattering of the magnons by defects and impurities is *enhanced*. Also the number of magnons acting as scattering sources for the conduction electrons is reduced. As a consequence a weaker temperature dependence of the magnon thermal conductivity is observed. In HoIn_3 the scattering of magnons by impurities overwhelms the effects of their increase in number, and therefore a maximum in the magnon thermal conductivity is seen.

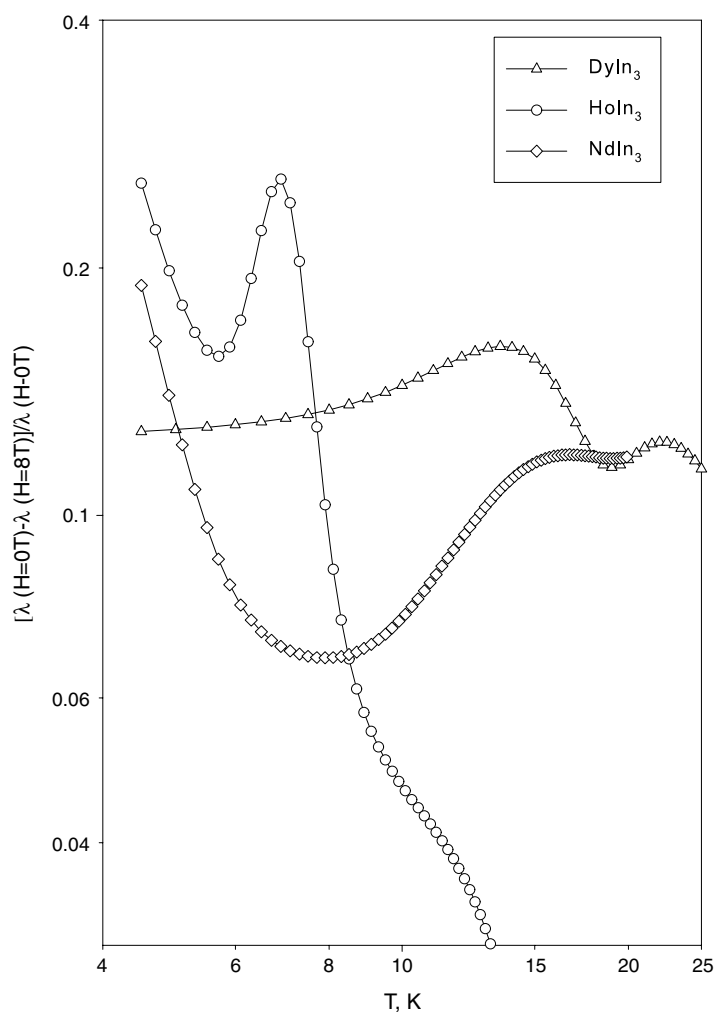


Figure 7. The temperature dependence of the magnetotransport: $\{[\lambda(T)_{H=0T} - \lambda(T)_{H=8T}]/\lambda(T)_{H=0T}\}$.

3. Summary

In the paper we present first results of the measurements of the thermal conductivity of REIn₃ compounds (RE = Pr, Nd, Dy, Ho, Tm) in the temperature interval 4–300 K in the absence of an external magnetic field. Thorough examination of the dependences found was made in the paramagnetic phase ($T > T_N$) and in the vicinity of and below their Néel temperatures T_N . In the latter case an external magnetic field of strength 8 T was applied in the direction parallel to the temperature gradient. In the paramagnetic phase we found the influence of the crystal field on the conduction electron scattering and the resultant conductivity; a rapid increase in the thermal conductivity is observed in the vicinity of the temperature in this phase. Below the Néel temperature an additional magnon contribution to the thermal conductivity was separated out. A separation of other apparent contributions seems impossible because of a lack of theoretical models describing the phenomena. The obtained results indicate that the magnetic field lowers

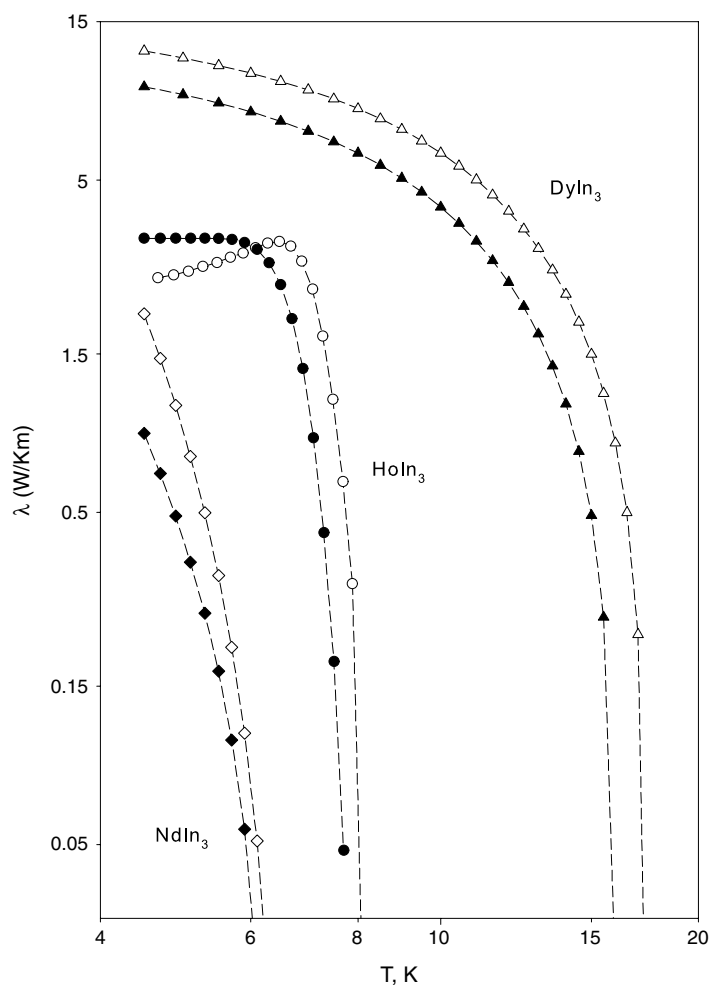


Figure 8. The magnon thermal conductivity of DyIn_3 , HoIn_3 and NdIn_3 as a function of the temperature.

the Néel temperature (the minimum at the curve $\lambda(T)_{H=8T}$). It also considerably lowers the thermal conductivity (from several to about 20%; see figure 7). Examination of the Lorenz function showed that the conduction electron scattering is inelastic in the whole examined temperature interval in both the absence and the presence of an external magnetic field, though for HoIn_3 and NdIn_3 the external field has an influence on the values of $L(T)/L_0$. Finally the magnon heat transport was determined in the presence of an external field and in its absence and was analysed as a function of the temperature.

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