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## The electronic Lorenz number in some transition metals at high temperatures

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**Abstract.** The usual method of plotting the high-temperature electrical resistivity  $\rho$  and thermal conductivity  $\lambda$  data on transition metals in the form of  $\lambda T$  against  $T^2/\rho$  plots gives a negative lattice thermal conductivity  $\lambda_g$ , when applied to V, Rh, Pd and Co. It is shown here that  $\lambda T$  for these metals, plotted against  $T^2$ , gives positive  $\lambda_g$  and electronic Lorenz numbers  $L_e^*$ , which depend on  $T$  in a particular manner, namely in proportion to  $\rho$ . These values of  $L_e^*$  broadly conform to Kolomoets' theory.

When applied to the data on titanium, zirconium and hafnium, the values obtained for  $L_e^*$  tend, at the highest temperatures, to the temperature-independent values of  $L_e$ , obtained from the  $\lambda T$  against  $T^2/\rho$  plots. In the case of polycrystalline and wire-form niobium, the values of  $L_e^*$  conform to Kolomoets' theory while the  $L_e$  obtained from the  $\lambda T$  against  $T^2/\rho$  plots do not.

### 1. Introduction

The electronic Lorenz number  $L_e$  of a normal metal, defined by the ratio of  $\rho$  to  $w_e T$  (where  $\rho$  and  $w_e$  are, respectively, the electrical and electronic thermal resistivities at the absolute temperature  $T$ ) should asymptotically approach the normal Sommerfeld value  $L_n (= 2.45 \times 10^{-8} \text{ V}^2 \text{ K}^{-2})$  at high temperatures  $T \gg \theta_D$  (Makinson 1938).

It was predicted by Kolomoets (1966) and demonstrated recently (Sharma *et al* 1986) that the characteristic electronic Lorenz number of a transition metal at high temperatures should, generally speaking, be greater or smaller than  $L_n$ , according to whether the d band is nearly empty or nearly full, or, in other words, according to whether the parameter  $\gamma = (E_F - E_0)/kT$  is positive or negative. Here  $E_F$  is the Fermi energy and  $E_0$  that of the proximate band edge;  $k$  is the Boltzmann constant. In the special case where  $\gamma$  is positive but less than 1.5,  $L_e$  should be smaller than  $L_n$ .

Attention was also drawn to the correlation between the sign of the deviation  $\Delta L_e (= L_e - L_n)$  of the high-temperature electronic Lorenz number from  $L_n$  and that of the deviation from linearity (DFL) of the high-temperature electrical resistivity of a transition metal.

Vanadium, rhodium, palladium and cobalt have defied all attempts at an analysis of their high-temperature electrical and thermal conductivity data to obtain the electronic Lorenz number  $L_e$  and to separate the electronic component  $\lambda_e$ , the lattice component

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$\lambda_g$  and (in the case of ferromagnetic cobalt) the magnon component  $\lambda_m$  of the measured thermal conductivity  $\lambda$ .

We present here a possible, essentially phenomenological method for obtaining this information in a limited temperature range.

## 2. The method of analysis

Since the probability of phonon umklapp scattering at high temperatures  $T \gg \theta_D$  is proportional to  $T$ , we can assume  $\lambda_g$  under these conditions to be inversely proportional to  $T$  (Peierls 1929, Liebfried and Schlomann 1954). Also, by definition,  $\lambda_e = L_e T / \rho$ , so that we may write

$$\lambda = \lambda_e + \lambda_g = L_e T / \rho + A / T$$

or

$$\lambda T = L_e T^2 / \rho + A \quad (1)$$

where  $A$  is a numerical constant.  $\lambda T$  plotted against  $T^2 / \rho$  gives a straight line for many of the transition metals (Sharma *et al* 1986). The gradient of the straight line directly gives (a constant)  $L_e$  at high temperatures for each transition metal, while  $\lambda_g$  is obtained by dividing the intercept on the  $\lambda T$  axis by  $T$ .

Such a plot made from the available data for each of the metals V, Ph, Pd and Co gives a negative intercept on the  $\lambda T$  axis. This means a negative  $\lambda_g$  and is unacceptable.

We have now used the same data but plotted  $\lambda T$  against  $T^2$ . In each case, if the data pertain to a wide temperature range, the result is a slightly curved line, concave to the  $T^2$  axis. However, a straight line results, with a positive intercept on the  $\lambda T$  axis, provided that we consider restricted temperature ranges, 1000–1700 K for V, 1000–1300 K for Pd and 1450–2050 K for Rh.

It can be seen that the present method presumes that  $L_e / \rho$  (or, in other words,  $\lambda_e / T$ ) has a constant value for each of these metals in the limited temperature range mentioned. Let us see whether this is justifiable.

From the free-electron theory of metals,  $\lambda_e = \frac{1}{3} C_e \bar{v} l$ , where  $C_e$  is the electronic specific heat,  $\bar{v}$  is the mean electron velocity and  $l$  the electron mean free path. At high temperatures  $T > \theta_D$ , for an electron gas subject to Fermi–Dirac statistics,  $C_e$  is proportional to  $T$ ,  $\bar{v}$  is constant and  $l$  is proportional to  $T^{-1}$ .  $\lambda_e$  (and also  $L_e$ , since  $\rho / T$  is constant for a normal metal at high temperatures) should therefore be independent of  $T$ . Since  $\lambda_g$  is expected (Peierls 1929) to be inversely proportional to  $T$  at these temperatures, the total  $\lambda$  should fall with increasing  $T$ . In fact,  $\lambda$  for these metals increases (more or less linearly) with increasing  $T$ , so that  $\lambda_e$  rising roughly in proportion to  $T$  is quite plausible in these metals—which implies that  $L_e$  varies roughly in proportion to  $\rho$ .

## 3. Discussion of the results

### 3.1. Vanadium, rhodium and palladium

We consider here the data of Voronin *et al* (1970) on vanadium above 1000 K, as an example, for the application of the present method of analysis (table 1).

**Table 1.** Data of Voronin *et al* (1970) on vanadium and the results of the application of the present method of analysis.

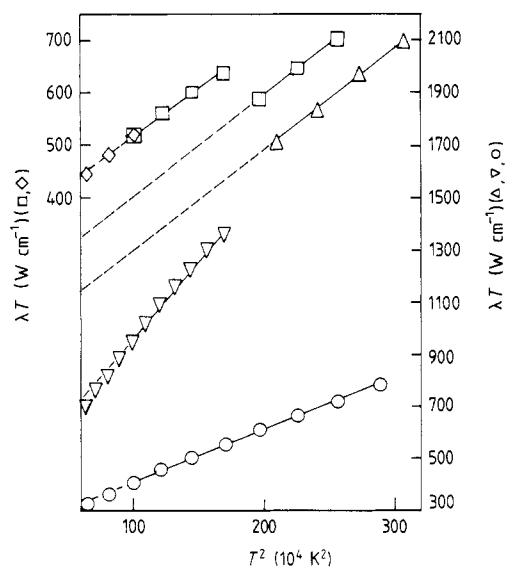
Data of Voronin <i>et al.</i>			Results of the present analysis			
$T$ (K)	$\rho$ ( $\mu\Omega$ cm)	$\lambda$ ( $W$ cm $^{-1}$ K $^{-1}$ )	$\lambda_g = 200/T$ ( $W$ cm $^{-1}$ K $^{-1}$ )	$\lambda_c = \lambda - \lambda_g$ ( $W$ cm $^{-1}$ K $^{-1}$ )	$\lambda_c/T$ ( $W$ cm $^{-1}$ K $^{-2}$ )	$10^8 L_e^*$ ( $V^2$ K $^{-2}$ )
1000	64.0	0.405	0.200	0.205	0.205	1.31
1100	69.1	0.412	0.182	0.230	0.209	1.42
1200	74.7	0.420	0.167	0.253	0.211	1.53
1300	79.0	0.427	0.154	0.274	0.211	1.62
1400	83.3	0.435	0.143	0.292	0.209	1.71
1500	87.9	0.442	0.133	0.305	0.203	1.80
1600	92.0	0.450	0.125	0.325	0.203	1.89
1700	97.0	0.457	0.118	0.340	0.200	2.00

The slope of the straight line of  $\lambda T$  against  $T^2$  is  $0.205 \times 10^{-3} W$  cm $^{-1}$  K $^{-2}$ . This gives  $L_e/\rho$ , so that  $L_e^*$  at any temperature is obtained by multiplying this slope by the corresponding value of  $\rho$ . The asterisk indicates the unusual temperature dependence, in proportion to  $\rho$ , in a restricted temperature range. The intercept on the  $\lambda T$  axis, when divided by  $T$ , gives the corresponding  $\lambda_g$ . Table 1, sixth column, shows that  $(\lambda - \lambda_g)/T = \lambda_c/T$  is constant within  $\pm 3\%$ .

We have tried numerous alternative procedures:

- (i)  $L_e = L_n$ ;
- (ii)  $L_e$  values read off from Makinson's curve;
- (iii)  $L_e$  having a constant value less than  $L_n$ , e.g.  $2.2 \times 10^{-8} V^2 K^{-2}$ ;
- (iv)  $L_e$  having a constant value greater than  $L_n$ , e.g.  $2.6 \times 10^{-8} V^2 K^{-2}$ .

None of these gives a  $\lambda_g$  which is theoretically acceptable.



**Figure 1.**  $\lambda T$  plotted against  $T^2$ :  $\circ$ , vanadium, data of Voronin *et al* (1970);  $\Delta$ , rhodium, data of Sorokin *et al* (1969);  $\nabla$ , palladium, data of Laubitz and Matsumura (1972);  $\square$ , cobalt, data of Zinovyev *et al* (1968), above 1000 K;  $\diamond$ , cobalt, data of Montague *et al* (1979), below 1000 K.

Table 2. Results of the present method of analysis.

Metal	Data source for $\rho$ and $\lambda$	Temperature range (K)	Sign of DFL of high temperature $\rho$ from table 3	Highest temperature for which $\lambda_g/\lambda$ and $L_c$ are tabulated (K)	$\lambda_g/\lambda$	$10^8 L_c^*$ ( $V^2 K^{-2}$ )	Sign of $L_c - L_n$	Conformity with Kolomoets' theory
V	Voronin <i>et al</i> (1970)	1000-1700	-	1700	0.26	2.0	-	Yes
Rh	Sorokin <i>et al</i> (1969)	1450-2050	+	2050	0.36	1.85	-	Yes, if $\gamma < 1.5$
Pd	Laubitz and Matsumura (1972)	1000-1300	-	1300	0.22	2.2	-	Yes
Co	Zinovyev <i>et al</i> (1968)	1000-1600	-	1600	0.31	1.88	-	Yes

The high value of  $\lambda_g$ , which is comparable with  $\lambda_e$ , of a metal such as V, whose Fermi surface is located just after a peak of the curve of the electronic density of states against the electron energy, could be a consequence of the high probability that s electrons are trapped in the holes in the d band, so that they are unable to limit the phonon mean free paths.

The results obtained from the present method of analysis based on  $\lambda T$  against  $T^2$  plots for V, Rh, Pd and Co are shown in figure 1 and summarised in table 2.

The DFL of the high-temperature electrical resistivity of a transition metal is due to the strong energy dependence of the density of states in the d band near the Fermi level. It can be positive or negative depending on the location of the Fermi level on the density-of-states curve (Jones 1956). The Fermi surfaces of V, Pd and Co are known to be situated just beyond peaks of the 3d, 4d and 3d density-of-states curves (Nemnonov 1965). Table 3 shows the DFL of the high-temperature electrical resistivity to be negative for these three metals. They are said to belong to the negative group.

**Table 3.** Sign of DFL of the high temperature electrical resistivity of V, Rh, Pd and Co.

Metal	$T_2$ (K)	$\rho_2$ ( $\mu\Omega$ cm)	$T_1$ (K)	$\rho_1$ ( $\mu\Omega$ cm)	$(\rho_2/T_2)/(\rho_1/T_1)$	Sign of DFL of $\rho$ at high temperatures
V	1700	97.0	1000	64.0	0.89	–
Rh	2050	48.8	1450	32.4	1.065	+
Pd	1300	38.09	1000	31.92	0.92	–
Co	1600	94.0	1400	85.0	0.97	–

Data sources for  $\rho$  as in table 2.

The signs of  $d\chi/dT$  and  $dC/dt$  of transition metals at high temperatures ( $\chi$  and  $C$  being the magnetic susceptibility and specific heat, respectively) are known (Shimizu *et al* 1962) to conform to the DFL of their  $\rho$  at high temperatures. Since the specific heat of rhodium (Shimizu and Katsuki 1964) did not vary with the temperature, it was suggested that the increasing and decreasing tendencies (as  $T$  rises) mutually cancel out. From this, it was inferred that the Fermi surface of rhodium lies half-way up the slope of the 4d density-of-states curve. Although table 3 shows rhodium to have a positive DFL, it can be entirely accounted for by thermal expansion and the corresponding decrease in  $\theta_D$  with rise of temperature—as in the noble metals. On the other hand, table 2 shows  $L_e^*$  at 2050 K (which is close to the melting point) to be less than  $L_n$ . According to Kolomoets, this means that the Fermi surface of rhodium should lie just after the minimum in the density-of-states curve, with  $\gamma < 1.5$ .

### 3.2. Cobalt

In a ferromagnetic metal under a temperature gradient (and owing to its magnetisation gradient  $d\mu/dz$ ), exchange interaction results in a flow of up-spin electrons in the direction  $\text{grad } \mu$  and a flow of down-spin electrons in the opposite direction. This component of the thermal conductivity is denoted as  $\lambda_{\text{spin}}$  or  $\lambda_{\text{magnon}}$  (or  $\lambda_m$ ) (Petrova 1963). It vanishes at the transition from the ferromagnetic to the paramagnetic state and accounts for the sharp break in the temperature dependence of the thermal conductivity of a ferromagnetic metal at the Curie point.

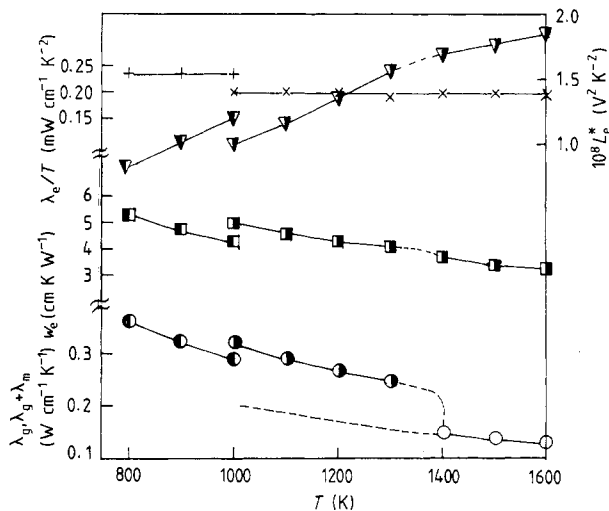


Figure 2.  $L_e^*$  ( $\nabla$ ),  $\lambda_g + \lambda_m$  ( $\bullet$ ),  $w_e$  ( $\blacksquare$ ),  $\lambda_e/T$  ( $\times$ ) and  $\lambda_g$  ( $\circ$ ), obtained by the present analysis for cobalt from the data of Zinovyev *et al* (1968):  $\nabla$ ,  $\bullet$ ,  $\blacksquare$ ,  $\times$ , corresponding  $L_e^*$ ,  $\lambda_g + \lambda_m$ ,  $w_e$  and  $\lambda_e/T$  data from Montague *et al* (1979) (no values of  $\lambda_g$  were determined by Montague *et al* since their data do not extend into the paramagnetic temperature region).

Figure 1 presents the thermal conductivity data of Zinovyev *et al* (1968) for cobalt from 1000 to 1600 K in the form of a  $\lambda T$  against  $T^2$  plot. Two straight lines (of nearly the same gradients) are drawn through the data points: one for the paramagnetic and the other for the ferromagnetic temperature region. The intercept of the former on the  $\lambda T$  axis, divided by the values of  $T$  in the paramagnetic region, gives  $\lambda_g$  at those temperatures. Similarly the intercept of the latter straight line on the  $\lambda T$  axis, divided by the values of  $T$  in the ferromagnetic region, gives  $\lambda_g + \lambda_m$  (shown at the bottom of figure 2). It is assumed that  $\lambda_m$ , like  $\lambda_g$ , has an inverse temperature dependence (Petrova 1963).

The slope of each straight line, multiplied by the value of  $\rho$  at any particular temperature in the region, gives  $L_e^*$  at that temperature. The data of Montague *et al* (1979) for cobalt below 1000 K are also included in figures 1 and 2 for comparison.

For ferromagnetic cobalt, for which  $\lambda$  decreases as  $T$  rises, the  $\lambda T$  against  $T^2$  plot is a straight line with a positive slope, implying that  $\lambda_e$  increases in proportion to  $T$ . As the dominant component  $\lambda_g + \lambda_m$  is inversely proportional to  $T$ , this becomes possible because the decrease in  $\lambda$  is slower than inversely proportional to  $T$ , enabling  $\lambda T$  to rise as  $T$  increases.

Figure 2 presents  $L_e^*$ ,  $\lambda_g$ ,  $\lambda_g + \lambda_m$ ,  $w_e$  and  $\lambda_e/T$  for cobalt as functions of  $T$ , based on the data from the two sources mentioned above. There is a slight bend in the lines for  $L_e^*$ ,  $\lambda_e$  and  $w_e$  at the Curie temperature. Phonon scattering limits the electronic thermal conductivity  $\lambda_e$  in the paramagnetic region. In the ferromagnetic region, while magnons serve as additional carriers for heat transport, they also provide an additional mechanism for scattering the conduction electrons. Hence the curve for  $w_e$  rises at the transition from the paramagnetic to the ferromagnetic state.

The Fermi surface of cobalt is situated just beyond a peak in the curve of the density of states against the electron energy (Nemnonov 1965). According to Kolomoets' theory,  $L_e$  should be less than  $L_n$ —as is indeed seen to be the case from figure 2 and table 2.

The polytherms for  $L_e^*$  and  $\lambda_e/T$  for cobalt below 1000 K based on the data of Montague *et al* (1979) are somewhat higher, and those for  $\lambda_g + \lambda_m$  and  $w_e$  based on the same data lie somewhat lower, than expected from the corresponding polytherms at temperatures above 1000 K based on the data of Zinovyev *et al* (1968).

One of the reasons could be as follows. As mentioned in § 2, the points at the bottom

end of the  $\lambda T$  against  $T^2$  plots for vanadium and palladium present a negative deviation from linearity and this deviation rapidly (numerically) increases at lower temperatures. The data of Montague *et al* (1979) for cobalt below 1000 K demonstrate this point, when compared with the data of Zinovyev *et al* (1968) above 1000 K. The former can be considered to be linear, but with a steeper slope (and hence a larger  $L_e^*$ ) and a smaller intercept on the  $\lambda T$  axis (and hence a smaller  $\lambda_g$ ) than would result from an extension of the straight line passing through the data points above 1000 K.

### 3.3. Titanium, zirconium and hafnium

In the case of V, Rh, Pd and Co, we had to resort to  $\lambda T$  against  $T^2$  plots since the more conventional  $\lambda T$  against  $T^2/\rho$  plots gave a negative  $\lambda_g$ . The latter method is applicable to Ti, Zr and Hf (in which  $\lambda$  again increases with increasing  $T$  at high temperatures) as it gives a positive  $\lambda_g$  varying in inverse proportion to  $T$  at high temperatures. So, we have made  $\lambda T$  against  $T^2$  plots (which also give a positive  $\lambda_g$  inversely proportional to  $T$ ) in order to check whether the results obtained by the two methods are mutually compatible.

Figure 3 presents the values of  $L_e^*$  and  $L_e$  for Ti, Zr and Hf. In each case, it can be seen that the temperature-independent  $L_e$  (obtained from the  $\lambda T$  against  $T^2/\rho$  plots) serve as the limits to which the values of  $L_e^*$  (obtained from the same data plotted in the form  $\lambda T$  against  $T^2$ ) tend at the highest temperatures.

Above 900 K,  $L_e^*$  (and also  $L_e$ ) for Ti and Hf are all larger than  $L_n$ , conforming to Kolomoet's rule for a nearly empty d band with  $\gamma > 1.5$ .

Zinovyev and Geld (1972) and Zinovyev *et al* (1972) suggest the presence of minor groups of electronic states near the Brillouin zone boundary, to account for the reduced slope of the  $\rho$ - $T$  curves for Ti, Zr, Hf and Y at high temperatures. For such small groups of carriers having an energy of the order of (or even less than)  $kT$ , the number of carriers interacting with the phonons becomes exhausted with increasing  $T$ , and any extra contribution arising from this mechanism would 'saturate'.

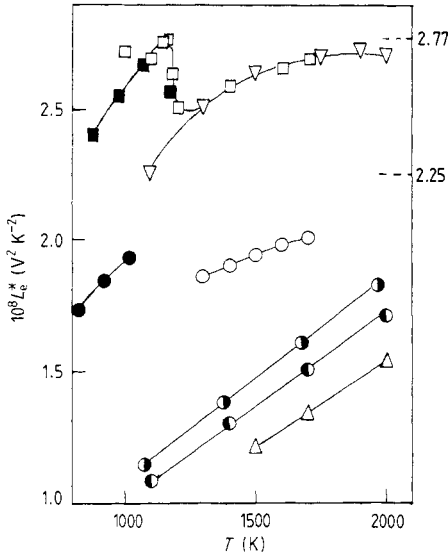
Mooij (1973) used the word 'saturation' for alloys having at least 20% admixture, with a temperature coefficient of resistance given by  $\alpha = (1/R) dR/dT < 100 \text{ ppm K}^{-1}$ , between 25 and 75 °C. We are dealing with unalloyed transition metals with  $\alpha \approx 500 \text{ ppm K}^{-1}$  for V between 1500 and 1700 K, 760  $\text{ppm K}^{-1}$  for Rh, 435  $\text{ppm K}^{-1}$  for Co and 500 for Pd between 1200 and 1300 K. The existing theories of 'saturation' have to be developed further before they can be employed in the present context.

### 3.4. Niobium

High-temperature conductivity data of Makarenko *et al* (1970) for niobium in polycrystalline, single-crystal and wire forms were analysed by us (Sharma *et al* 1986) by plotting  $\lambda T$  against  $T^2/\rho$ . The values thus obtained for  $L_e$  were, respectively,  $2.4 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$ ,  $2.6 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$  and  $2.5 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$ . Since the Fermi surface of niobium is known to be situated just after the maximum in the curve of density of states against electron energy,  $L_e$  is expected to be smaller than  $L_n$ , following Kolomoets' rule.

We have now drawn  $\lambda T$  against  $T^2$  plots for the same data (and also the data of Neimark and Voronin (1968)) for Nb. The values of  $L_e^*$  thus obtained are seen (figure 3) to conform to Kolomoets' rule, being much less than  $L_n$ , even at 2400 K (the figure could not accommodate temperatures above 2000 K). It appears to us that the present method should be adopted when the straight line in the  $\lambda T$  against  $T^2/\rho$  plot passes





**Figure 3.** Values of  $L_e^*$ : ■, titanium, data of Shamashov *et al* (1971); □, titanium, data of Arutyunov *et al* (1971); ●, zirconium, data of Powell and Tye (1961); ○, zirconium, data of Peletskii *et al* (1970); ▽, hafnium, data of Arutyunov *et al* (1972); ○, niobium, data of Makarenko *et al* (1970), single-crystal form; △, niobium, data of Makarenko *et al* (1970), wire form (the results for niobium in polycrystalline form were omitted to avoid confusion); ●, niobium, data of Neimark and Voronin (1968); --- (on right), temperature-independent values  $L_e$  for hafnium and titanium ( $2.77 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$ ) and for zirconium ( $2.25 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$ ) obtained from  $\lambda T$  against  $T^2/\rho$  plots.  $L_e^*$  for niobium turns out to be smaller than  $L_n$  for all the three forms, agreeing with Kolomoets' theory.

through (or very close) to the coordinate origin, as in the case of Nb. Since there can be a scatter of as much as  $\pm 5\text{--}10\%$  in the measured values of  $\lambda$  at high temperatures, one cannot even be sure of the sign of  $\lambda_g$ , unless it is of significant magnitude.

Further, since the transition metals are rather poor conductors, one cannot expect  $\lambda_e$  to dominate to such an extent as in normal good conducting metals. For the single crystal, polycrystal and wire,  $\lambda_g/\lambda$  at 2000 K comes out to be 0.35, 0.32 and 0.38 (whereas the earlier method gave 0.03, 0.07 and even less). These are roughly of the same magnitude as those shown in table 2 for V, Rh, Pd and Co.

We refer, in passing, to the electrical and thermal conductivity data on niobium for temperatures up to 1300 K ( $\rho$  up to 1600 K) reported by Moore *et al* (1980). For the same reason as given above, a  $\lambda T$  against  $T^2$  plot is preferred here also. For temperatures above 800 K, this gives a straight line with a slope of  $3.5 \times 10^{-4} \text{ W cm}^{-1} \text{ K}^{-2}$ , which gives  $L_e^* = 1.79 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$  at 1300 K and  $\lambda_g/\lambda = 0.3$ . As explained earlier,  $\lambda T$  against  $T^2$  plots at lower temperatures result in a higher  $L_e^*/\rho$  and a lower  $\lambda_g$  than the plots based on the data at higher temperatures.

#### 4. Conclusions

According to the free-electron theory of metals, the electronic thermal conductivity  $\lambda_e$  should be independent of  $T$ , at temperatures  $T \gg \theta_D$ . Since, for a normal non-transition metal,  $\rho/T$  is constant at high temperatures, their product (which, by definition, is the electronic Lorenz number  $L_e$ ) should then be constant. For a transition metal at such temperatures, it was predicted by Kolomoets (1966) and shown by Sharma *et al* (1986) that  $L_e \geq L_n$ , correlating with the positive or negative deviation of  $\rho/T$  (at high temperatures) from constancy. A temperature dependence of  $L_e$  is not specifically envisaged in a transition metal at high temperatures in Kolomoets' theory.

In a transition metal, in which the thermal conductivity is found to increase with increasing  $T$  at high temperatures, the proportionality of  $\lambda_e$  to  $T$  was initially a conjecture,

but it has now been shown to be true from our analysis of the conductivity data on V, Rh, Pd, Co, Ti, Zr, Hf and Nb (see table 1 for example of V). An  $L_e$ -value varying in proportion to  $\rho$  (even though in a limited temperature region) follows from this by virtue of the definition  $L_e = \lambda_e \rho / T$ .

A  $\lambda T$  against  $T^2$  plot should be preferred not only when the  $\lambda T$  against  $T^2/\rho$  plot gives a negative intercept on the  $\lambda T$  axis (as in the case of V, Rh, Pd, Co, etc) but also when the intercept is so close to zero that one is not sure that it is positive (as in the case of Nb).

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