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Thermopower due to defects in Ni, Pd and Pt

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Abstract

The thermopower induced by tensile plastic deformation at a near-room temperature is measured to determine the change in thermopower per unit dislocation density $\Delta S_d/N_d$ in nickel (99.98%). The dislocation density is determined indirectly by measurement of the flow stress. The partial-wave method is used to calculate the contribution of dislocations and vacancies to the thermoelectric power of nickel, platinum and palladium. The value $\Delta S_d/N_d$ calculated for nickel agrees quite satisfactorily with measured ones. These results are the first quantitative estimation of the defect contribution to thermopower of transition metals.

1. Introduction

Among the electron-transport properties, thermoelectric power (TEP) is the most sensitive and informative but at the same time the least understood one. For example, there are plenty of experimental and theoretical data on the electrical resistivity change caused by various structural defects including measurements per unit density, and a scarcity of such data for the thermoelectric power.

The thermoelectric properties of noble metals are most investigated from the viewpoint of its relation to defects. It has been established that dislocations raise, whereas vacancies lower, the absolute thermopower of these metals. The ratios of thermopower to electrical resistivity induced by vacancies, $\Delta S_v/\Delta\rho_v$ [1, 2], and dislocations, $\Delta S_d/\Delta\rho_d$ [2], have been measured. The thermopower change per unit concentration of vacancies [1, 2] and dislocations [3] has been determined; the characteristic thermopower induced by dislocations has been estimated [4]. The results of measurements are in good correlation with the calculated contribution of vacancies and dislocations to the thermopower of noble metals [5–7].

The transition metals are investigated much less. For example, Polak investigated the extra-thermoelectric power of platinum after quenching and after plastic deformation in temperature from -190 to 20°C [2]. It was found that dislocations and vacancies reduce the absolute value of thermopower. Available experimental results for nickel [8–10] allow us to make a conclusion only about a positive sign of the thermoelectric effect induced by plastic deformation. Since the demagnetization of samples was not carried out in these experiments, the authors' attempt to

separate a component connected with vacancies seems not to be credible, as the thermoelectric effect induced by deformation is masked by the magnetic hysteresis of the thermoelectric effect. The magnetic hysteresis of the thermoelectric effect on pure nickel was investigated in the work [11]. Besides, in the articles [8–10] the change of thermopower was not connected with the density of the defects, that complicated the theoretical interpretation of the results.

In this work, we present the experimental evaluation of the extra thermopower per unit of dislocation density ($\Delta S_d/N_d$) for nickel on the basis of direct measurement of the thermopower induced by plastic deformation and indirect estimation of the dislocation density. The measured value is compared with the theoretical one calculated by the partial-wave method. The calculations are performed both for nickel and the nickel sub-group (Pd and Pt). The dislocation model used here was applied earlier for calculation of the dislocation residual resistivity of a wide range of metals, including transition ones [12].

2. Experiment and results

The measurement technique is similar to that used earlier for measurement of the dislocation contribution to the thermopower of copper, silver and zinc [3]; however, it was complemented by some features connected with tuning out from the influence of ferromagnetic ordering in nickel. We measured the thermopower induced by plastic tensile deformation of polycrystalline wire samples of nickel (99.98%) 0.4 and 0.5 mm in diameter. The samples had a V-like shape with a 17–18 cm shoulder. To remove the defects and stresses induced upon preparation, the samples were annealed at a pressure of 10^{-2} Pa for 3 h at 1000 °C, and then furnace-cooled to room temperature. One branch of the sample was fixed, and the other was subjected to tensile deformation with a continuously increasing load being cyclically applied and removed until the neck origin. The maximum plastic deformation was 30–35%. The measurements were carried out at room temperature. The absolute thermopower of our nickel samples in annealed state at room temperature was found to be -19.8 ± 0.1 , that coincides with the available experimental data lying in the range from -19.24 to $-20.0 \mu\text{V K}^{-1}$ [13]. A constant temperature difference of 50 ± 0.5 °C was maintained along the sample with a heat controller. The deformation-induced thermoelectric voltage was measured to an accuracy of 2×10^{-8} V. The sample elongation was detected accurate to 0.2 mm.

Because nickel at near-room temperature is a ferromagnet with negative magnetostriction, its tension causes not only elastic and plastic deformation, but also magnetization of the sample perpendicular to the applied load, owing to reorientation of the magnetic moments of domains along the direction of easy magnetization. It creates an additional contribution to the thermopower (a thermoelastic effect). After removal of the load, besides the TEP induced by plastic deformation, the residual thermoelectric effect caused by irreversible change of domain structure remains (hysteresis of thermoelectric effect). Therefore, at each step of unloading, the sample is demagnetized in variable magnetic field with amplitude decreasing to zero. The results of the measurement are shown in figure 1. Each point represents a value that was averaged over four samples.

Curve 1 shows the variation of the TEP in the deformed sample branch with respect to the branch that is undeformed in the loaded state; curve 2 in the unloaded state; curve 3 after demagnetization in the unloaded state. Curve 3 represents the thermopower induced by structural defects that arise during plastic deformation (dislocations and point defects). We do not carry on annealing of point defects. However, earlier obtained results for pure nickel which was subjected to various methods of cold working at near-room temperature [14, 15] showed that point defects (mainly vacancies) are annealed in a temperature range of 80–250 °C, removing an approximately one-third increment of the electrical resistance induced by cold

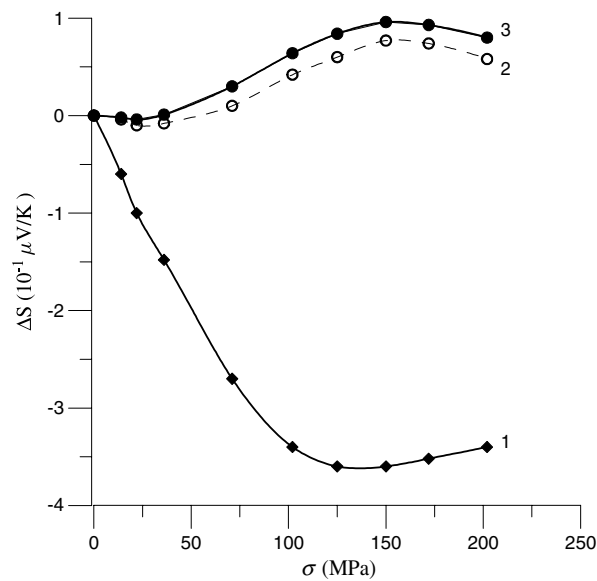


Figure 1. Dependence of the tension-induced thermopower on the applied tensile stress for nickel: 1—in the loaded state of the sample; 2—in the unloaded state; 3—after demagnetizing in the unloaded state.

working. The remaining increment of the electrical resistance is caused by dislocations and annealed at higher temperatures (550–650 °C) [14]. Thus, one can believe that with an accuracy of 30–35% all of the change in curve 3 can only be attributed to dislocations. Therefore, we can state that the sign of the dislocation-induced thermoelectric effect in nickel is positive; that is, the dislocations reduce the negative absolute thermoelectric power of nickel. This conclusion about the sign of the effect correlates with the results of the works [8, 9].

As known, the defect structure of metal determines unambiguously the macroscopic flow stress through the mechanism of work hardening. Between the flow stress σ and the average dislocation density N_d a simplest correlation was found [16–18],

$$\sigma \simeq \frac{1}{2}GbN_d^{1/2}, \quad (1)$$

where G is the shear modulus, b is the Burgers vector, and the stress σ is considered as the resolved shear stress that is required for further deformation. This relationship was established on the basis of electron-microscopic studies for both pure metals and solid solutions [16, 17], and was grounded theoretically [18]. This formula was used in a number of earlier studies for quantitative estimation of the increment in the electrical resistivity per unit dislocation density $\Delta\rho_d/N_d$ [19], and the results agreed satisfactorily with more accurate and more recent results based on direct measurements of dislocation density. In this work the average dislocation density N_d was determined from equation (1) on the basis of curve 3 in the quasi-linear part (area of intense hardening).

Since our samples had a drawing texture caused by their preparation technique and subsequent tensile deformation, in a first approximation the value of σ was taken to be the tensile stress σ_{tens} . Taking the tabular data $G = 75$ GPa [20] as the shear modulus and the Burgers vector of a stable perfect dislocation $a_0\sqrt{2}/2$ for FCC metals (where a_0 is the lattice parameter) as b , we obtain the average change of the thermopower per unit dislocation density $\Delta S_d/N_d = (4.3 \pm 1.5) \times 10^{-18}$ V cm² K⁻¹.

Table 1. Input and intermediate parameters of calculation.

Metal	a_0 (Å) [40]	b (Å)	k_F (Å ⁻¹)	E_F (eV)	Δx_F	ρ_0 ($\mu\Omega$ cm) [13]	S_0 (μ V K ⁻¹) [13]
Ni	3.52	2.49	1.135	4.91	-5.8	6.20	-17.83
Pd	3.89	2.75	1.034	4.08	-3.7	9.71	-9.70
Pt	3.92	2.77	0.937	3.35	-10.0	9.80	-4.29

3. Calculation of thermoelectric characteristics of defects

In view of the absence in the literature of any quantitative data on the contribution of dislocations and vacancies to thermopower of nickel, we try to calculate these characteristics, both for the nickel, and for other transition metals of the nickel subgroup (Pd and Pt). The transport properties of transition metals containing defects are described on the basis of Mott's theory [21]. The main feature of electronic structure of these metals is that they have two overlapping bands (s and d) with $10(s + d)$ electrons per atom near the Fermi energy. As the effective mass of d holes considerably exceeds the effective mass of s electrons, the d holes do not take part in the transfer processes; however, they are effective scatterers of s electrons because of the high density of electron states in the d band and consequently the high probability of s-d transitions. Here it is assumed that the number of electrons with spin up in the s band is equal to the number of electrons with spin down, and the scattering of s electrons in vacant states of the d zone occurs without spin overturn.

The calculation is carried out by the partial-wave method. The model of dislocation used here was tested earlier for calculation of dislocation residual resistivity in nickel and other transition metals and has shown quite good agreement with experiment [12]. We believe that the dislocation core brings in the basic contribution to the electron scattering. The influence of the long-range elastic fields surrounding the dislocation is neglected, as it is ten times weaker [22].

As a scattering potential of the dislocation we used an axially symmetrical potential of the form [12]

$$V(r) = \begin{cases} V_1, & r < R_1 \\ V_0, & R_1 \leq r \leq R_2 \\ 0, & r > R_2. \end{cases} \quad (2)$$

Such potential causes the repulsion of the electrons by surplus negative charge due to dilatation of the lattice and the possibility of the existence of the resonance quasi-stationary electron states. The electron states on dislocations near the Fermi energy have been discussed in the works [23–25, 12]. Here we believe the resonance level coincides with the Fermi energy, $E_r = E_F$. This condition determines the depth of the potential well V_1 .

The dilatation value in the dislocation core is taken as $\Delta V = b^2$ per unit of dislocation length [26]. The external radius of the potential is determined from the dilatation value ($\Delta V = \pi R_2^2$) and coincides with the atomic one. The used values b are given in table 1 for the metals under study. The internal radius R_1 is a free parameter; it is set as $R_1 = R_2/2$. As shown in [7], the scattering cross section and residual resistivity of dislocation are weakly sensitive to the choice of the parameter. Below it will be shown that calculated additional thermopower per unit dislocation density is insensitive to the choice of the radius R_1 as well if the height of the potential barrier V_0 is determined self-consistently from the Fridel screening

Table 2. Residual resistivity and additional thermopower of vacancies and dislocation for transition metals.

Metal	$(\Delta x_Q)_v$	$\Delta\rho_v/c_v$ ($\mu\Omega$ cm/at.%)	$\Delta S_v/c_v$ (μ V/(K at.%)	$(\Delta x_Q)_d$	$\Delta\rho_d/N_d$ (10^{-19} Ω cm ³)	$\Delta S_d/N_d$ (10^{-18} V cm ² K ⁻¹)
Ni	0.71	2.94	11.73	-0.77	5.43	2.43
Pd	0.65	3.24	4.89	-0.78	7.20	1.25
Pt	0.56	3.36	7.90	-0.66	9.02	2.36

condition [12]

$$\frac{k_F}{\pi^2} \int_0^\pi \sum_{m=-\infty}^{\infty} \eta_m(\varphi) \sin \varphi \, d\varphi = \xi, \quad (3)$$

where ξ is the line density of charge along the dislocation axis, $\xi = n_s \Delta V / \Omega_{at}$, Ω_{at} is the atomic volume, n_s is the number of carriers per atom, and η_m is the m th phase shift of the asymptotic solution of the Schrödinger equation. The solution of the Schrödinger equation for the potential (2) is given in [12] in detail. The values n_s are equal to 0.54, 0.55 and 0.42 e/atom for Ni, Pd and Pt respectively [27, 12]. These values coincide with the average atomic magnetic moments, as the d bands in the metals are filled and the number of electrons in the s band is equal to the number of holes in the d band.

The spherical potential barrier of atomic radius and height V_0 is used as a scattering potential for vacancies. The height of the potential barrier is determined from the screening condition, which for the point defects has the form [28]

$$Z = \frac{2}{\pi} \sum_{l=0}^{\infty} (2l+1) \eta_l, \quad (4)$$

where Z is the surplus charge of the defect ($Z = -n_s$), l is the orbital quantum number, and η_l is the l th phase shift. The analytical expression for the phase shift is conventional [29].

The transport scattering cross section Q for defects of spherical symmetry is written as

$$Q = \frac{4\pi}{k_F^2} \sum_{l=0}^{\infty} (l+1) \sin^2(\eta_l - \eta_{l+1}), \quad (5)$$

where k_F is the wavevector at the Fermi level, on the basis of the free-electron approximation $k_F = (3\pi^2 n_s / \Omega_{at})^{1/3}$, and Ω_{at} is atomic volume. The numerical values of the wavevectors k_F and the Fermi energy E_F in the free-electron model are given in table 1.

The increase of electrical resistivity due to the atomic portion c_i of vacancies is calculated as

$$\Delta\rho_v = \frac{\hbar c_i Q k_F}{n_s^*}, \quad (6)$$

where n_s^* is the effective number of carriers per atom which take part in scattering on defects. For the metals under study $n_s^* = n_s/2$, and it is 0.27, 0.275 and 0.21 e/atom for Ni, Pd and Pt correspondingly [27, 12]. Here we keep in mind that only half of the electrons take part in scattering on defects; the other half with opposite spin direction scatters into vacant states of the d band. The concentration of vacancies expressed as an atomic percentage is $c_v = 100c_i$.

The calculated residual resistivity of vacancies per atom per cent $\Delta\rho_v/c_v$ is presented in table 2. We can compare these values with available experimental data. So, according to various sources the vacancies in platinum contribute to resistivity (5.75 ± 03) $\mu\Omega$ cm/at.% [30] and 2.4 $\mu\Omega$ cm/at.% [31] (our calculated value is 3.36 $\mu\Omega$ cm/at.%). The measured magnitude

of $\Delta\rho_v/c_v$ for nickel constitutes $5 \mu\Omega \text{ cm/at.}\%$ [27] and $3.2 \mu\Omega \text{ cm/at.}\%$ [32] (the calculated value is $2.94 \mu\Omega \text{ cm/at.}\%$). We did not consider the lattice relaxation near the vacancy. However, in accordance with the works [33, 34] this effect is insignificant and constitutes from 3 to 10%. Thus one can say that there is a quite good correlation between the calculated values of $\Delta\rho_v/c_v$ and the available experimental data.

The change of thermopower per unit defect concentration (additional thermopower) $\Delta S_i/N_i$ is found from the relation [35]

$$\frac{\Delta S_i}{N_i} = S_0 \frac{\Delta\rho_i}{\rho N_i} \left(\frac{\Delta x}{x} - 1 \right) \quad (7)$$

where $\rho = \rho_0 + \Delta\rho_i$,

$$x = - \left(\frac{d \ln \rho_0}{d \ln E} \right)_{E_F};$$

$$\Delta x = - \left(\frac{d \ln \Delta\rho_i}{d \ln E} \right)_{E_F} = - \frac{k_F}{2Q} \left(\frac{dQ}{dk} \right)_{k_F} + \left(\frac{d \ln F}{d \ln E} \right)_{E_F} = \Delta x_Q + \Delta x_F;$$

here S_0 and ρ_0 are the absolute thermopower and electrical resistivity of annealed metal at temperature T ; k_B is the Boltzmann constant; F is the area of the Fermi surface; and E_F is the Fermi energy. For the point defects $N_i = c_i/\Omega_{\text{at}}$. These expressions are valid when it is possible to neglect the phonon-drag effect and inelastic scattering by defects.

The parameter $\Delta x_F \equiv V$ is determined on the basis of experimental data on the thermoelectric size effect, from which the value $U = (d \ln \lambda / d \ln E)_{E_F}$ can be found using the expression

$$S_0 = - \frac{\pi^2 k_B^2 T}{3e E_F} \left(\frac{d \ln \lambda}{d \ln E} + \frac{d \ln F}{d \ln E} \right)_{E_F} = - \frac{\pi^2 k_B^2 T}{3e E_F} (U + V), \quad (8)$$

where e is the magnitude of electron charge and λ is the free path. For example, for nickel the value $U = 19$ [36]. Measurements performed for platinum defined the amount $(d \ln \lambda / d \ln E)_{E_F} = (3.63 \pm 0.83) \text{ eV}^{-1}$ [37], which at $E_F = 3.35 \text{ eV}$ (here) gives $U = 12.2$. For palladium, one adduces the results of measuring the quantity $\pi^2 k_B^2 T V / 3e E_F = -6.2 \mu\text{V K}^{-1}$ at the temperature $T = 280 \text{ K}$ [38]; putting $E_F = 4.08 \text{ eV}$ results in $V = -3.7$. Used in our calculation values $V = \Delta x_F$, and also S_0 and ρ_0 for the temperature $T = 273 \text{ K}$ are presented in table 1. The calculated additional thermopower and residual resistivity due to vacancies and dislocations are given in table 2. The dislocation residual resistivity $\Delta\rho_d/N_d$ coincides with our earlier results [12].

The change of the thermopower per unit of dislocation density $\Delta S_i/N_i$ should not depend substantially on the temperature for considered metals, as S_0 and ρ_0 have practically linear dependence on the temperature in the range of validity the equations (7).

As shown in [7], the change of the radius R_1 of the potential (2) did not substantially influence the calculated electrical resistivity of dislocations $\Delta\rho_d/N_d$, but gave rise to an essential change in the thermoelectric parameter Δx_Q . Nevertheless, the additional dislocation thermopower $\Delta S_d/N_d$ is found to be sufficiently stable to choose the radius R_1 . So, the variation of the ratio $n = R_2/R_1$ in the range from 0.1 to 0.9 results in a change of the dislocation resistivity $\Delta\rho_d/N_d$ by no more than 7.5%, and the value $\Delta S_d/N_d$ by less than 4% for nickel. For palladium, the values are 6.5% and 2%, respectively.

4. Discussion

As follows from table 2, where results of the calculation are presented, the vacancies and dislocations bring in a same sign positive contribution to the TEP of metals under study. This

correlates with results of measurements carried out on platinum by Polak [2]. The positive contribution of dislocations to the thermopower of nickel obtained in the works [8, 9] is justified in our measurements too. The opposite sign contribution of vacancies to the TEP of nickel in comparison with dislocations discovered in the works [9, 10] seems not to be convincing enough, as already noted above. We did not find the experimental data on the contribution of vacancies or dislocations to the thermopower and electrical resistivity of palladium.

The value of $\Delta S_d/N_d$ calculated here is $2.43 \times 10^{-18} \text{ V cm}^2 \text{ K}^{-1}$, that agrees quite satisfactorily with the measured value $(4.3 \pm 1.5) \times 10^{-18} \text{ V cm}^2 \text{ K}^{-1}$, without taking into account the contribution of point defects (mainly of vacancies).

It is possible to roughly estimate the vacancy contribution to the thermopower induced by plastic deformation from our measurements on the basis of the calculated value of $\Delta S_v/c_v$ for nickel, if we evaluate the vacancy concentration from the relation $c_i \approx 10^{-4}\varepsilon$ [39] (where ε is the value of the relative plastic deformation). So, the tension-induced TEP $\Delta S \approx 0.09 \mu\text{V K}^{-1}$ corresponds to the relative plastic deformation $\varepsilon \approx 0.25$. The vacancy concentration corresponding to this deformation is $c_v = 0.25 \times 10^{-2} \text{ at.}\%$. In accordance with our calculation, such vacancy concentration in nickel should result in a change in thermopower by the value of $\Delta S_v \approx 0.03 \mu\text{V K}^{-1}$. If we subtract this value from ΔS and connect the remaining part with the dislocation density by formula (1), then we obtain the change of thermopower per unit dislocation density in nickel $\Delta S_d/N_d \approx (2.4 \pm 0.8) \times 10^{-18} \text{ V cm}^2 \text{ K}^{-1}$, which is in better accordance with the results of our calculation.

It should be noted that the experimental values of $\Delta S_d/N_d$ presented here are slightly underestimated, since the true resolved shear stress σ is equal to the average projection of the tensile stress σ_{tms} on the slip direction. If we assume that the sample texture is such that the directions of easy slip in different grains make an angle θ no more than $\pi/4$ with the tensile direction (which seems to be most probable), then we have $\sigma = 0.85\sigma_{\text{tms}}$, as a result of solid-angle averaging. In this case, the measured value $\Delta S_d/N_d$ will be 1.4 times more.

5. Conclusions

Thus, summing up, one could say that the additional dislocation thermopower in nickel, calculated on the basis of the earlier proposed dislocation model, shows a quite satisfactory agreement with the measured value, both in value and in sign of the effect, in spite of the rough evaluation of the dislocation density induced by plastic deformation. The value and the positive sign of the thermoelectric effect connected with vacancies in nickel represent only the theoretical conclusion, and require experimental verification. The results of the calculation for platinum agree with available experiment in sign; however, the numerical data also need experimental confirmation, as the results for Pd.

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