International Journal of Thermophysics, Vol. 2, No. 4, 1981

The Influence of Neutron Irradiation on the Thermal Conductivity of Aluminum in the Range 5–50 K

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Received September 9, 1981

Measurements of thermal conductivity of 6N to 3N pure aluminum in the temperature range 5–50 K subjected to fast neutron irradiation, with exposures of 10^{13} and 10^{16} $n \cdot cm^{-2}$, are reported. The thermal conductivity maximum was found to shift towards higher temperatures with an increase in the fast neutron irradiation exposure. At high temperatures, a departure from Wilson's theory was observed, which may be attributed to the existence of additional electron scattering mechanisms. An increase in both ideal and residual thermal resistivity components with an increase in the radiation exposure was noted.

KEY WORDS: aluminum; low temperature; neutron irradiation; thermal conductivity.

1. INTRODUCTION

The irradiation of solids by different particles may lead to changes in the structure of the crystal lattice and, as a result, to changes in the transport properties. These changes may be due to generation of vacancies, interstital atoms, impurity atoms, or entire disordered regions. Fast neutron irradiation generates mainly point defects, clusters of point defects (Brinkman's areas [1]), and a single cluster of point defects.

According to the results reported in the literature [2], the physical defects in copper caused by fast neutron irradiation decreased the thermal conductivity significantly, up to a factor of 25, in the temperature range 4.5–100 K. In this paper, results of measurements on aluminum in the range 5–50 K subjected to fast neutron irradiation are reported.

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0195-928X/81/1200-0341$03.00/0

Table I. The Residual Resistivity Ratio (RRR) of the Aluminum Samples of Different Purities and Different Neutron Irradiation Exposures[a]

Sample	Residual resistivity ratio 6N	 5N	 4N	 3N
Unirradiated	3392	1504	223	96
irradiated, 10^{13} n · cm^{-2}	1615	971	144	63
irradiated, 10^{16} n · cm^{-2}	1208	893	121	52

[a] Data from ref. [3].

2. EXPERIMENTS

2.1. Samples

The samples investigated were aluminum wires 3 mm in diameter and about 75 mm long with purities in the range 6N to 3N. They were annealed at 540°C for 4 h in an inert gas atmosphere [3]. The samples were irradiated with fast neutrons (about 1 MeV) to exposures of 10^{13} n · cm^{-2} and 10^{16} n · cm^{-2} in liquid nitrogen. During mounting in the cryostat, the samples were at room temperature for 2–4 h and then were cooled to the liquid nitrogen or liquid helium temperatures [3]. The chemical and physical purities of the investigated aluminum samples were characterized by the residual resistivity ratio (RRR) measurements. The RRR values for the aluminum samples are presented in Table I.

2.2. Method of Measurements

The thermal conductivity was measured by a steady-state axial heat flow method in the temperature range 5 to 50 K [3].[2] The aluminum samples were mounted in the experiment chamber presented in Fig. 1. The cylindrical chamber was made of a 0.5 mm thick copper sheet. The round copper plate 1, which was soldered to the cold block, was the upper part of the chamber. The copper block 2 that holds the sample was soldered to the plate. The upper heater 3, wound on the copper plate, was used to raise the mean temperature of the sample to the desired value. The heater 4, wound on a copper frame at the lower end of the sample, was for generating the temperature difference in the sample. In order to achieve better thermal contact, the lower heater was cemented to the sample with low-temperature glue.

The thermometer probes, 5 for the measurement of difference and 6 for the measurement of absolute temperature, were attached to the samples

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[2] For an explanation of symbols, see nomenclature at end of article.

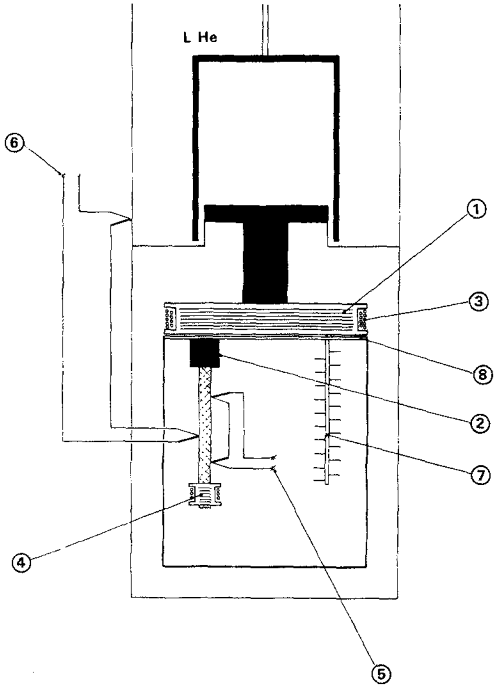

Fig. 1. Schematic diagram of the experiment chamber: 1, copper plate; 2, holder; 3, copper heater; 4, sample heater; 5, 6, thermometers; 7, heat exchanger; 8, chamber screen.

of 3N, 4N, and 5N purities, with copper holders. The temperature drop along the samples of 6N purity was measured in the range 5-50 K with a calibrated germanium thermometer having an uncertainty of 0.01-0.04 K.

2.3. Errors

The maximum relative error in the measured thermal conductivity for the 6N pure aluminum samples was about 2.5%. For the samples of purities in the range 5N to 3N (5.8, 34.4, 1700 ppm, respectively), the maximum relative error in the measured thermal conductivity varied from about 5% to about 2%.

3. RESULTS AND DISCUSSION

The variation of thermal conductivity as a function of temperature for both unirradiated and irradiated aluminum samples with purities in the range 6N-3N is shown in Figs. 2-5. The corresponding numerical data are

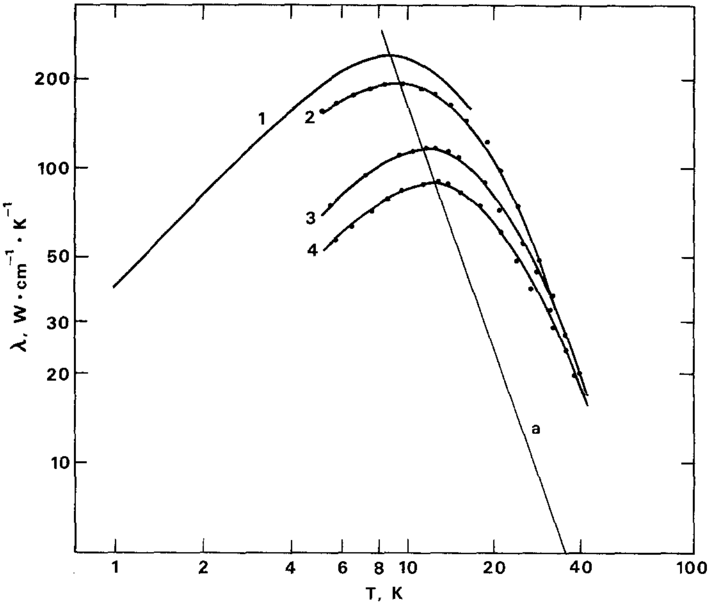

Fig. 2. Thermal conductivity vs temperature for 6N pure aluminum. 1, recommended TPRC (CINDAS) curve [4, 5]; 2, unirradiated; 3, exposure of 10^{13} n · cm^{-2}; 4, exposure of 10^{16} n · cm^{-2}.

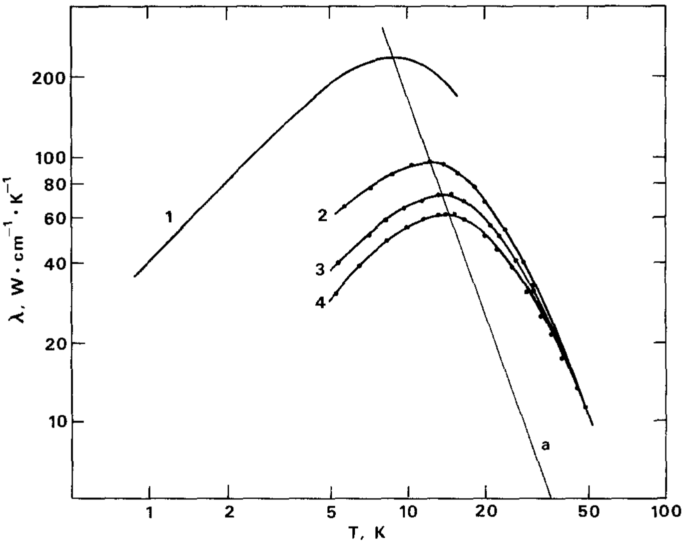

Fig. 3. Thermal conductivity vs temperature for 5N pure aluminum. 1, recommended TPRC (CINDAS) curve [4, 5]; 2, unirradiated; 3, exposure of 10^{13} n · cm^{-2}; 4, exposure of 10^{16} n · cm^{-2}.

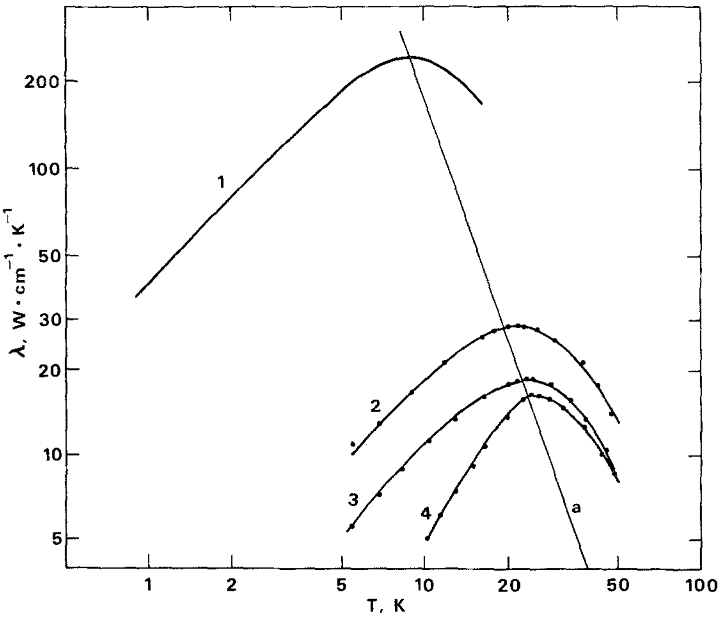

Fig. 4. Thermal conductivity vs temperature for 4N pure aluminum. 1, recommended TPRC (CINDAS) curve [4, 5]; 2, unirradiated; 3, exposure of 10^{13} n · cm^{-2}; 4, exposure of 10^{16} n · cm^{-2}.

presented in tabular form in the Appendix (Tables A1–A4). It was observed that the value of the maximum thermal conductivity, λ_m, decreased with increasing irradiation and the temperature, T_m, corresponding to the maximum thermal conductivity shifted toward higher temperatures. A similar variation was also noted for 5N pure copper [2] over the range 4.5 to 100 K irradiated with fast neutrons (exposures of 1.6×10^{19}, 4.3×10^{19}, and 6.5×10^{19} n · cm^{-2}).

The results of relative fractional change in thermal conductivity maximum, $\Delta\lambda_m/\lambda_m$, for irradiated aluminum samples and for copper samples [2] are presented in Table II. The largest percentage change in thermal conductivity maximum of aluminum for a given irradiation exposure is for the sample with 6N purity. This means that heat transport in pure aluminum is more sensitive to neutron irradiation than in impure aluminum. The values of the residual thermal resistivity coefficient β both for irradiated aluminum of this work and irradiated copper [2] are given in Table III.

The thermal conductivity results were correlated with the use of the equation given in ref. [6], which is based on the modifications of the simple relation for thermal conductivity given by Wilson [7]. According to the

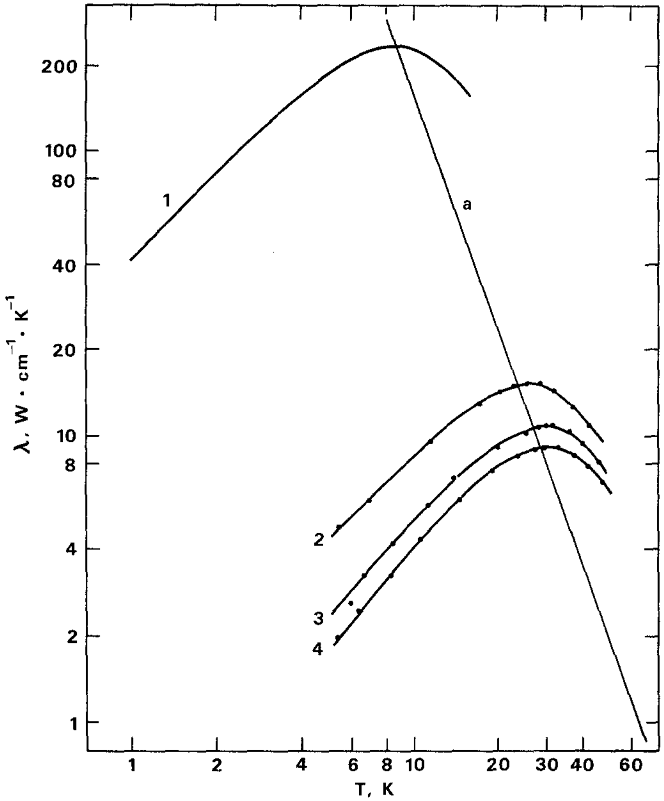

Fig. 5. Thermal conductivity vs temperature for 3N pure aluminum. 1, recommended TPRC (CINDAS) curve [4, 5]; 2, unirradiated; 3, exposure of 10^{13} n · cm^{-2}; 4, exposure of 10^{16} n · cm^{-2}.

Table II. Relative Percentage Change in Thermal Conductivity Maximum ($100 \cdot \Delta\lambda_m/\lambda_m$) for Aluminum and Copper of Different Purities for Different Irradiation Exposures

Material	Exposure (n · cm^{-2})	$\frac{\Delta\lambda_m}{\lambda_m} \cdot 100\%$			
		6N	5N	4N	3N
Aluminum	10^{13}	39	24	32	29
	10^{16}	53	35	42	39
Copper[a]	1.6×10^{19}		75		
	4.3×10^{19}		83		
	6.5×10^{19}		88		

[a] Values estimated from the data given in ref. [2].

Table III. Values of the Coefficient β both for Aluminum and Copper of 5N Purity for Various Neutron Irradiation Exposures

Material	Exposure $(\mathrm{n \cdot cm^{-2}})$	β $(\mathrm{W^{-1} \cdot cm \cdot K^2})$
Aluminum	10^{13}	0.13
	10^{16}	0.16
Copper[a]	1.6×10^{19}	0.50
	4.3×10^{19}	1.18
	6.5×10^{19}	1.72

[a] Values estimated from the data given in ref. [2].

equation in ref. [6],

$$\lambda = \left(\alpha' T^n + \frac{\beta}{T}\right)^{-1} \tag{1}$$

where

$$\alpha' = \alpha''\left(\frac{\beta}{n\alpha''}\right)^{(m-n)/(m+1)} \tag{2}$$

The constants α'' and n are related to the ideal component of thermal resistivity, and $-m$ denotes the slope of the straight line that crosses the maximum values of thermal conductivity, λ_m. The coefficient β is associated with the residual thermal resistivity. For the aluminum samples with 6N and 5N purities (Figs. 2 and 3), the straight line a crossing λ_m values has a slope equal to $-m$ within the measurement uncertainties. However, in the case of less pure irradiated samples (4N, Fig. 4; and 3N, Fig. 5), the slope of the straight line that crosses λ_m values deviates from the value $-m$.

According to the relation given by Wilson [7], thermal resistivity may be written as follows:

$$W_e = \frac{1}{\lambda} = W_i + W_0 \tag{3}$$

where W_i is the ideal thermal resistivity associated with scattering of electrons by the thermal vibrations of the lattice ($W_i = \alpha T^2$) and W_0 is the residual thermal resistivity associated with the scattering of electrons by impurities and defects ($W_0 = \beta / T$).

After substituting the equivalents for W_i and W_0 in Eq. (3) and rearranging, one obtains

$$W_e T = \alpha T^3 + \beta \tag{4}$$

The coefficients α and β may be obtained graphically by plotting the quantity $W_e T$, (T/λ), as a function of T^3. The slope of the straight line fitted to the points is α, and the y intercept is β. The parameter β obtained by the above procedure agrees, within a few percent, with the value obtained from residual electrical resistivity data and the Lorenz number.

Variation of the quantity T/λ as a function of the quantity $T^3(I_5(\theta/T)/I_5(\infty))$ for the aluminum samples is presented in Figs. 6–9. The term $I_5(\theta/T)$ is the Debye integral of the fifth order which, in the limit of $(\theta/T) \rightarrow \infty$, has the value of 124.4. Temperature dependence of the Debye integral was taken from the literature; the Debye temperature (θ) for aluminum was taken to be 410 K [7]. It may be observed from Figs. 6–9 that, in the temperature range 5–20 K, agreement between the experimental data and the results based on Wilson's theory is satisfactory. However, at higher temperatures, the experimental results deviate from the theoretical predictions. This may be attributed to the existence of additional electron scattering mechanisms not accounted for by Eq. (4). The clusters of point defects generated by neutron irradiation of the samples may possibly be the cause of the additional scattering of electrons. For samples having 6N and 5N purities, the Umklapp processes, which are characterized by an expo-

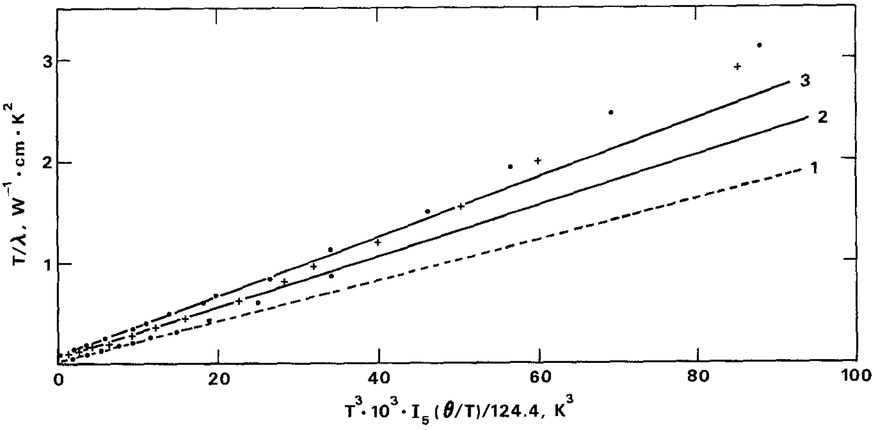

Fig. 6. The quantity T/λ vs $T^3(I_5(\theta/T)/124.4)$ for aluminum samples of 6N purity. 1, unirradiated; 2, exposure of 10^{13} n · cm^{-2}; 3, exposure of 10^{16} n · cm^{-2}.

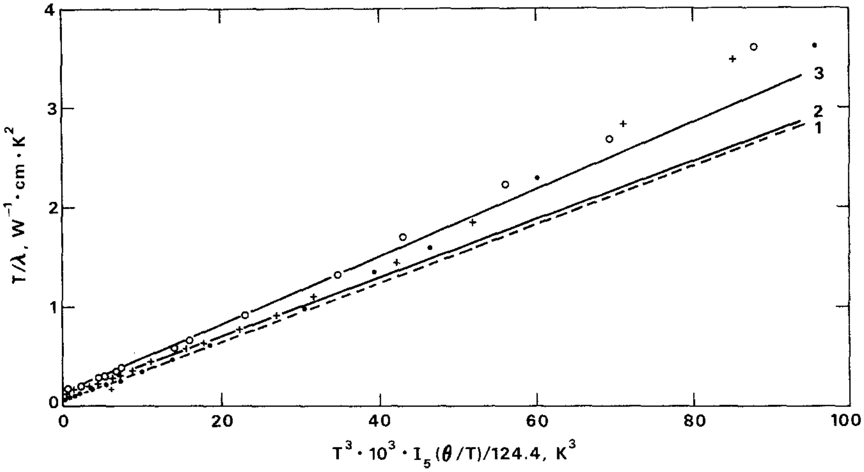

Fig. 7. The quantity T/λ vs $T^3(I_5(\theta/T)/124.4)$ for aluminum samples of 5N purity. 1, unirradiated; 2, exposure of 10^{13} n · cm^{-2}; 3, exposure of 10^{16} n · cm^{-2}.

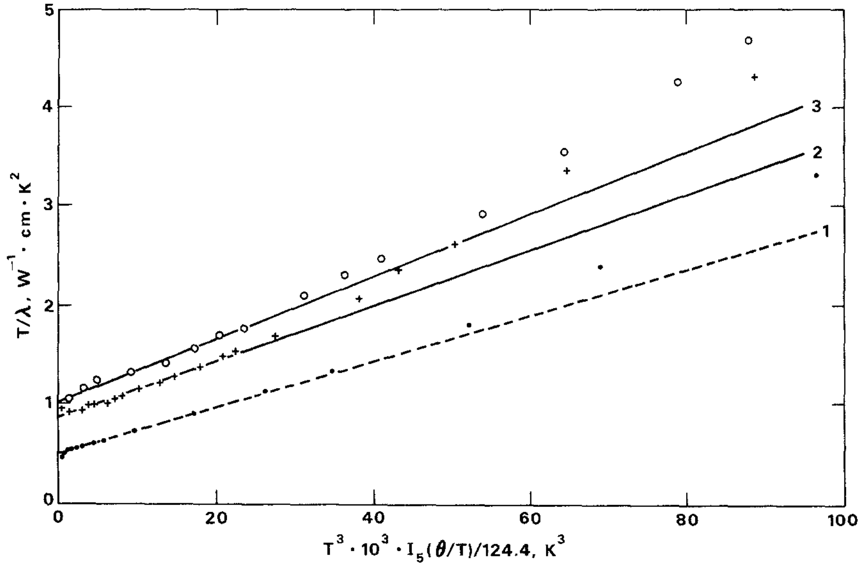

Fig. 8. The quantity T/λ vs $T^3(I_5(\theta/T)/124.4)$ for aluminum samples of 4N purity. 1, unirradiated; 2, exposure of 10^{13} n · cm^{-2}; 3, exposure of 10^{16} n · cm^{-2}.

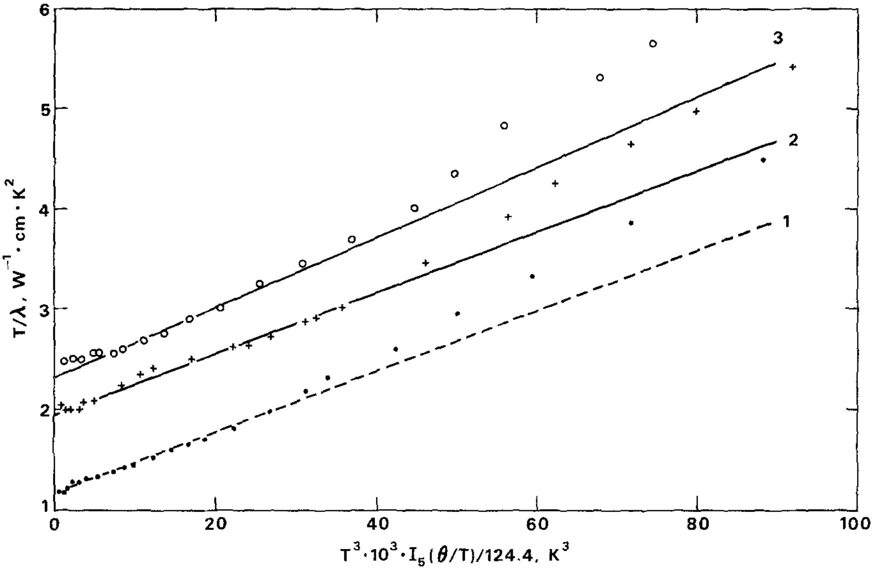

Fig. 9. The quantity T/λ vs $T^3(I_5(\theta/T)/124.4)$ for aluminum samples of 3N purity. 1, unirradiated; 2, exposure of 10^{13} n · cm^{-2}; 3, exposure of 10^{16} n · cm^{-2}.

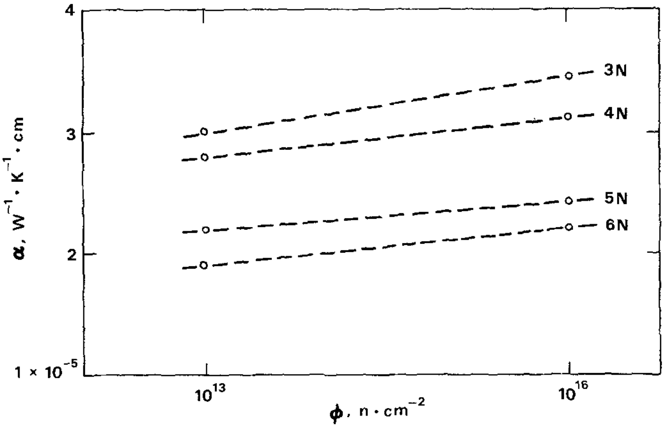

Fig. 10. The coefficient α vs neutron exposure for 6N to 3N pure aluminum.

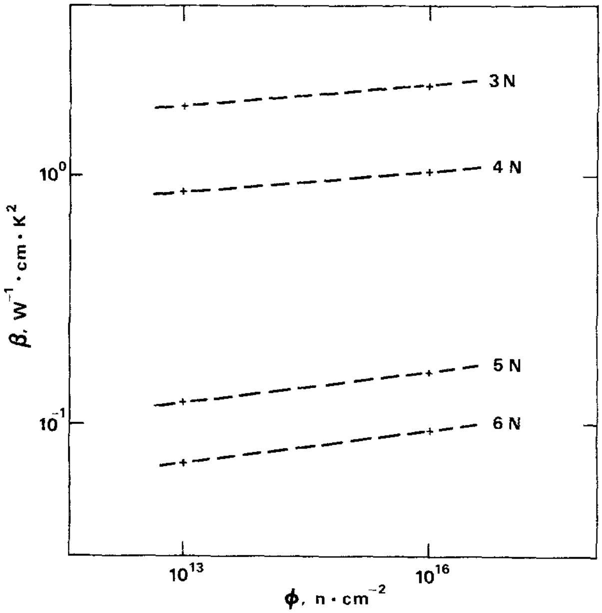

Fig. 11. The coefficient β vs neutron exposure for 6N to 3N pure aluminum.

nential dependence of the thermal resistivity on temperature, are also possibilities [8].

Dependence of the coefficient α on fast neutron irradiation exposure ϕ is shown in Fig. 10. It may be seen that the value of α increases with increasing irradiation exposure. Dependence of α on ϕ, indicating the influence of defects due to irradiation on the ideal thermal resistivity component, implies that the Matthiessen's rule [9] is not applicable to aluminum. Dependence of the coefficient β on fast neutron irradiation exposure is shown in Fig. 11. The increase in β with an increase in exposure is an evidence of the increase in the effective electron scattering by the defects generated by neutron irradiation.

At this time, dependences of both α and β for aluminum on irradiation exposure should be considered on a qualitative basis only. For a quantitative description of $\alpha(\phi)$ and $\beta(\phi)$, new experiments with several other neutron irradiation exposures have to be performed.

4. CONCLUSIONS

Thermal conductivity of the aluminum samples with 6N and 5N purities subjected to fast neutron irradiation follow the theoretically predicted pattern. Thermal conductivity maxima fall on a straight line as shown in Figs. 2 and 3. Thermal conductivity of the aluminum samples with 4N and 3N purities subjected to fast neutron irradiation shows a different pattern. Thermal conductivity maxima do not fall on a straight line as shown in Figs. 4 and 5. For the case of high exposures, a shift of maximum thermal conductivity toward higher temperatures is observed. Thermal conductivity of the aluminum samples with an increased impurity content and increased irradiation exposure shows an increased departure from the values predicted by the Wilson theory (Figs. 6–9).

The values of both of the coefficients α and β in Eq. (4) increase with increasing neutron irradiation exposure (Figs. 10 and 11). This means that an increase in scattering of electrons, both by thermal vibrations of the lattice and by defects generated by irradiation, takes place.

NOMENCLATURE

$I_5(\theta/T)$	Debye integral of the fifth order
$-m$	slope of the straight line that crosses maximum thermal conductivity values
n	exponent in ideal thermal resistivity component
T_m	temperature corresponding to maximum thermal conductivity
W_e	total electronic thermal resistivity
W_i	ideal thermal resistivity
W_0	residual thermal resistivity
α	ideal thermal resistivity coefficient in Eq. (4)
α'	ideal thermal resistivity coefficient in Eq. (1)
α''	constant related to the ideal part of thermal resistivity in Eq. (2)
$\alpha(\phi)$	ideal thermal resistivity coefficient depending on irradiation exposure
$\beta(\phi)$	residual thermal resistivity coefficient depending on irradiation exposure
λ	thermal conductivity
λ_m	maximum thermal conductivity
θ	Debye characteristic temperature
ϕ	irradiation exposure

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APPENDIX: TABLES A1, A2, A3, AND A4

Table A1. Thermal Conductivity, λ, of the 6N Pure Aluminum Sample

Unirradiated		Irradiated, 10^{13} n · cm^{-2}		Irradiated, 10^{16} n · cm^{-2}	
T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})
5.01	155.97	5.38	76.00	5.70	57.07
5.69	166.13	6.08	81.10	6.52	63.81
6.47	178.27	7.10	91.50	7.58	71.95
6.96	181.68	8.00	102.37	8.50	78.00
7.08	182.70	8.60	106.15	9.62	84.17
8.75	191.84	9.55	112.42	10.84	89.24
9.18	192.73	10.43	115.03	11.37	89.54
9.56	193.91	11.61	116.80	11.91	89.81
10.55	190.06	12.60	116.98	12.82	89.96
11.28	188.21	13.80	115.10	13.90	89.54
12.45	178.55	15.12	109.00	15.40	82.14
13.18	173.52	16.00	98.24	16.50	80.05
14.11	166.96	18.50	90.05	18.00	74.47
15.17	155.04	20.01	80.00	21.20	61.85
16.01	146.46	20.89	72.15	22.30	56.50
16.93	137.18	23.00	64.02	24.00	49.03
17.79	127.84	25.17	55.09	26.20	43.50
18.77	120.41	28.24	44.92	27.00	39.04
19.90	109.54	30.50	37.18	29.85	35.01
21.15	98.45	31.80	33.00	32.50	28.72
22.65	85.96	34.25	28.70	36.00	24.01
23.55	81.79	37.05	24.10	38.50	20.00
24.49	74.98	40.03	20.01	42.00	17.03
26.62	61.73	45.00	15.45	45.42	14.60
29.28	48.63				
32.53	36.84				

Table A2. Thermal Conductivity, λ, of the 5N Pure Aluminum Sample

Unirradiated		Irradiated, 10^{13} n · cm^{-2}		Irradiated, 10^{16} n · cm^{-2}	
T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})
5.61	67.12	5.30	40.86	5.28	30.52
6.20	72.08	6.12	45.48	5.80	35.01
7.01	77.85	7.06	51.30	6.43	39.12
8.12	85.32	7.20	54.42	7.71	44.00
8.60	89.57	8.10	59.12	8.20	49.14
9.52	90.04	9.60	65.04	9.90	55.72
10.25	95.16	11.22	69.92	11.55	59.80
11.00	95.41	11.79	73.00	12.72	61.82
12.08	97.73	13.01	73.89	13.93	62.05
12.65	97.42	13.60	74.06	14.56	62.12
13.50	96.00	14.50	73.94	15.00	62.09
15.25	89.67	16.10	69.91	16.25	60.44
17.50	79.52	18.20	65.21	17.45	57.03
19.40	69.15	19.36	63.90	18.98	54.11
21.50	62.00	20.62	56.74	19.42	50.33
24.00	50.10	22.21	50.24	22.02	45.50
26.50	43.20	24.90	45.08	25.10	37.51
30.00	33.05	26.00	41.50	28.50	31.04
34.20	25.26	28.17	36.43	32.75	25.00
36.10	22.50	30.05	33.00	35.30	21.50
40.08	17.50	31.70	29.20	39.20	17.47
43.17	14.98	35.00	24.50	39.20	17.47
46.50	12.75	37.50	20.10	42.04	15.54
		40.06	17.24	45.50	12.62
		42.40	15.02		
		45.05	12.97		

Table A3. Thermal Conductivity, λ, of the 4N Pure Aluminum Sample

Unirradiated		Irradiated, 10^{13} n · cm^{-2}		Irradiated, 10^{16} n · cm^{-2}	
T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})
5.50	11.20	5.50	5.71	6.10	2.55
6.20	12.00	6.00	6.60	7.34	3.15
6.95	13.08	6.95	7.30	8.50	4.02
7.70	14.90	7.75	8.65	9.55	4.64
8.80	16.66	8.25	9.08	10.35	5.13
9.00	17.00	9.50	10.43	11.15	5.94
9.90	18.60	10.40	11.28	12.20	6.68
11.05	20.00	11.95	12.72	12.80	7.25
11.70	21.09	12.80	13.62	13.85	8.11
13.00	23.07	13.90	14.64	15.15	9.31
14.50	25.03	15.45	15.47	16.45	10.87
16.05	26.60	16.30	16.24	18.75	12.22
17.60	27.63	17.95	17.50	21.45	14.74
19.60	28.20	18.85	17.56	23.75	16.84
21.25	29.04	19.85	18.22	25.80	16.53
22.25	28.95	21.55	18.58	27.35	16.07
25.75	28.04	23.15	18.78	28.65	16.24
28.00	27.00	24.25	18.76	31.60	14.95
29.65	26.00	26.00	18.50	33.20	14.32
31.40	20.70	27.35	18.46	34.60	14.02
32.72	24.50	28.10	18.20	37.90	12.93
37.60	21.56	30.20	17.80	41.00	11.50
42.00	17.60	33.75	16.16	43.85	10.30
47.00	14.20	35.20	15.00	45.50	9.76
		37.00	14.08		
		41.00	12.20		
		45.55	10.61		

Table A4. Thermal Conductivity, λ, of the 3N Pure Aluminum Sample

Unirradiated		Irradiated, 10^{13} n · cm^{-2}		Irradiated, 10^{16} n · cm^{-2}	
T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})	T (K)	λ (W · cm^{-1} · K^{-1})
5.50	4.75	5.40	2.30	5.40	1.95
6.20	5.31	6.00	2.60	6.38	2.46
7.00	5.89	6.80	3.22	7.40	2.72
7.80	6.50	7.50	3.54	8.50	3.25
8.10	7.31	8.56	4.21	9.50	3.71
9.90	8.28	9.00	4.95	10.70	4.34
10.40	8.70	10.45	5.25	11.30	4.65
11.60	9.61	11.40	5.70	13.00	5.19
13.25	10.42	12.80	6.40	14.82	5.94
14.27	11.15	14.10	7.15	15.80	6.41
15.75	12.01	15.35	7.50	16.73	6.50
17.38	13.10	16.90	8.12	17.50	6.85
19.20	13.82	19.27	8.44	19.30	7.55
20.50	14.30	20.05	8.98	20.23	7.78
21.25	14.65	22.00	9.40	22.21	8.30
23.00	15.04	23.00	9.60	23.75	8.59
24.40	15.25	25.50	10.25	25.52	8.87
25.50	15.38	28.00	10.75	27.30	9.04
26.60	15.49	28.80	11.01	29.35	9.06
28.10	15.32	30.05	11.05	31.50	9.14
29.90	15.11	31.50	11.07	33.42	9.08
31.58	14.45	32.00	11.05	35.60	8.92
32.50	14.00	33.00	11.01	36.85	8.51
35.00	13.50	36.00	10.48	39.10	8.18
37.00	12.60	39.24	10.15	41.70	7.95
39.90	12.05	40.52	9.60	43.05	7.74
42.20	11.00	42.50	9.24	46.00	7.28
45.50	10.25	44.00	8.96	47.50	7.08
		46.10	8.63		
		47.00	8.15		