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ABSTRACT

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The thermoelectric power of copper-aluminum (Cu-Al) and copper-silicon (Cu-Si) alloys has been determined from 4.2 to 320°K. Assuming that the thermopower is separable into diffusion and phonon-drag components, the change in phonon-drag thermopower ΔS_g is determined for Cu + 0.77 At.% Al and Cu + 1.12 At.% Si. The change in phonon-drag thermopower is analyzed using the method developed by Huebener for lattice vacancies in gold. Scattering of phonons by impurities is assumed to follow a Rayleigh scattering law with relaxation time $\tau_i = (a\omega^4)^{-1}$. From the change in phonon-drag thermopower it is found that $a = (0.9 \pm 0.4) \times 10^{-43}$ sec^3 for 0.77 At.% Al in Cu, and that $a = (6.5 \pm 2.3) \times 10^{-43}$ sec^3 for 1.12 At.% Si in Cu. These results are compared with the scattering parameter computed from the mass-difference term in Klemens' theory for scattering of low-frequency lattice waves by point imperfections. For Cu-Al, the results indicate that phonon scattering can be accounted for by the mass-difference term. The results for Cu-Si indicate that the mass-difference contribution to phonon scattering is of the same magnitude as the contribution from the combined effects of the elastic strain field and changes in the elastic constants of interatomic linkages.

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I. INTRODUCTION

Measurements of thermoelectric power have been previously used to study gross features of the Fermi surface (Ref. 1 and 2) as well as phonon-drag effects in copper based alloys (Ref. 3 and 4). In particular, Blatt and his co-workers carried out an extensive study of the phonon-drag thermopower in dilute copper alloys (Ref. 3 and 4). They separated the diffusion and phonon-drag components out of the thermopower, and observed quenching of the phonon-drag component by solute atoms of mass greater than copper. This Report is concerned with the change in phonon-drag thermopower due to solute atoms having mass smaller than that of the host copper atom. For this purpose, aluminum and silicon have been chosen as solute atoms.

The lattice thermal conductivity of Cu-Si alloys has been determined to 90°K by Klemens, White, and Tainsh (Ref. 5). They analyzed thermal conductivity in terms of phonon scattering (due to mass difference) using an approximate formula (Ref. 6 and 7) which gave reasonable agreement with experiment (Ref. 5). The agreement is considered surprising in view of the fact that lattice distortion is estimated to contribute approximately as much as the effect of mass difference to the scattering of lattice waves in the Cu-Si alloys (Ref. 5). The phonon-drag thermopower is also known to be sensitive to the scattering of phonons by these mechanisms. For example, using the change in phonon-drag thermopower due to lattice vacancies in gold, Huebener (Ref. 8) supplied evidence of phonon scattering due to the strain field associated with a lattice vacancy. In addition, Blatt has explicitly considered changes in the phonon-drag component due to large mass solutes in dilute copper alloys (Ref. 3 and 4). In this Report, the change in phonon-drag thermopower, with alloying, is used to obtain estimates of the separate contributions from mass difference and lattice distortion to phonon scattering. Of particular interest is the comparison of these results with the estimated contribution of the various scattering mechanisms in Cu-Si alloys (Ref. 5).

II. EXPERIMENTAL

A. Preparation of Specimens

Measurements of thermoelectric potentials were carried out on thermocouples, one leg of which was composed of high-purity copper and the other leg consisted of the alloy specimen. Pure copper wires were fabricated from American Smelting and Refining Company grade A-58 copper of 99.999% stated purity. This material was first melted under a dynamic vacuum of 3×10^{-6} torr. The resulting 0.4-in. D slug was swaged to 0.080 in. and drawn to the final diameter of 0.010 in. The wire was then vacuum annealed at 530°C for 24 hr. The sample was etched during fabrication and after annealing using a solution consisting of four parts H_2O and one part HNO_3. Spectrographic analysis after annealing showed no metallic impurities in addition to the impurities specified by the copper manufacturer. Gas analysis after fabrication revealed the following gaseous impurities in parts per million by weight: nitrogen 1.9, oxygen 12.8, and hydrogen 12.8.

Starting materials for the alloys, in addition to the copper mentioned above, were Super Raffinal[1] aluminum (5^9 purity), and Dow–Corning semiconductor grade silicon (6^9 purity). All materials were etched and weighed before combining in vacuum outgassed graphite crucibles. The melting operation was performed under a dynamic vacuum of 2×10^{-6} torr. The melt was shaken every 15 min over a period of 2 hr. To inhibit segregation, quenching followed the melting operation. The resulting billets were cold swaged from 0.4 to 0.080 in., and drawn to 0.010-in. D. Finally, the alloy wire was annealed at 600°C for 72 hr under a vacuum of 2×10^{-6} torr. Spectrographic and chemical analyses were performed on the finished product. In addition, the resistivities at 4.2, 77, and 273°K were measured. The solute content, as determined by spectrographic and chemical analyses, as well as the resistivities at liquid-helium temperatures, are shown in Table 1.

B. Measurements

The hot junction of the copper-alloy thermocouple was secured to a heater (mounted above the cryogenic fluid) with the cold junction immersed directly in liquid helium or liquid nitrogen depending on the temperature range covered. Measurements were carried out from 4.2 to 320°K. Voltages from the copper-alloy thermocouple were measured with a Rubicon model 2668 "thermofree" potentiometer, using a Guildline galvanometer amplifier as null

Table 1. Solute concentrations and residual resistivities of copper alloys

Solute atom	Concentrations At.%[a]	Residual resistivity μΩ/cm
Al	0.77	0.93
Al	2.37	2.89
Si	1.12	2.38
Si	2.68	6.54

[a] The pure copper had a residual resistivity of 2.53×10^{-3} μΩ/cm.

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[1] A.I.A.G. Metals Inc., New York, N. Y.

indicator. Germanium resistance thermometers were used to determine temperatures from 4.2 to 100°K, and copper-constantan thermocouples to determine temperatures above 100°K. Readings were taken every two degrees below liquid-nitrogen temperatures and at three degree intervals from 77 to 320°K. A least-squares fit of the emf vs T data was carried out using an IBM 1620 computer. Thermoelectric powers were then obtained from derivatives of the emf vs T data. Accuracy of the voltage measurements is ±0.01 μv, while temperatures were accurate to ±0.1°K. Resistivities were determined using the Rubicon potentiometer to measure voltage drop across the specimen. A Leeds and Northrup type K-3 potentiometer was used to determine current by measurement of voltage drop across a standard resistance in series with the current through the specimen.

III. SEPARATION OF PHONON-DRAG AND DIFFUSION COMPONENTS

The thermoelectric power of copper S^{Cu} is assumed to be the sum of the diffusion thermopower S_e^{Cu} and the phonon-drag component S_g^{Cu} (Ref. 3 and 4)

$$S^{Cu} = S_e^{Cu} + S_g^{Cu} \tag{1}$$

Experimentally the thermoelectric power of a thermocouple formed between pure copper and the alloy was determined. In this case

$$\Delta S = S^A - S^{Cu} = \Delta S_e + \Delta S_g \tag{2}$$

where S^A is the absolute thermopower of the alloy.

$$\Delta S_e = S_e^A - S_e^{Cu}$$
$$\Delta S_g = S_g^A - S_g^{Cu}$$

Where S_g^A is the phonon-drag component of the alloy thermopower and S_e^A the diffusion component, ΔS_g the quantity of interest can be obtained from ΔS provided the change in diffusion thermopower ΔS_e is known. The remainder of this Section is devoted to describing the method used in obtaining ΔS_e. (The method used here is similar to that employed in Ref. 4.)

Employing a variational method for solution of the Boltzmann equation, Kohler (Ref. 9) obtained the following relation for the diffusion thermopower S_e, where more than one contribution to S_e is present in a metal,

$$S_e = \frac{\sum_i W_e^i S_e^i}{\sum_i W_e^i} \tag{3}$$

where $\sum_i W_e^i$ is the electronic thermal resistivity of the metal, and W_e^i and S_e^i are contributions to the electronic thermal resistivity and diffusion thermopower due to the ith electron scattering mechanism.

Equation (3) is valid under the following conditions:

1. Electron scattering events in the metal can be treated in terms of a single homogeneous group of charge carriers.
2. The presence of impurities does not alter the Fermi surface.
3. The various electron scattering mechanisms act independently of each other.
4. The lattice heat conductivity is negligible; i.e., all of the effective heat should be transported by conduction electrons.

From Eq. (3), it follows that for a sufficiently dilute alloy

$$\Delta S_e = \frac{S_e^{Cu}}{\frac{W_e^0}{W_e^1} + 1}\left(\frac{S_e^1}{S_e^{Cu}} - 1\right) \tag{4}$$

where S_e^1 is the diffusion thermopower due to the solute, W_e^0 is the intrinsic electronic thermal resistivity of copper, and W_e^1 is the electronic thermal resistivity due to the solute atoms in copper; W_e^0 is obtained from existing data

(Ref. 10) on the thermal conductivity of high purity copper. At 320°K, a negligible phonon-drag contribution is assumed in the pure metal (Ref. 11) and, hence, in the alloy. The preceding statement signifies that the measured difference in thermopower between the alloy and pure copper at 320°K is, to a reasonable approximation, equal to ΔS_e. In addition, at 320°K $S^{Cu} \approx S_e^{Cu}$, and S_e^1 can be determined from Eq. (4) for this single temperature. Assuming that S_e^1 is proportional to temperature, it can then be determined by simple computation at other temperatures in the range covered (Ref. 4). It is thus readily seen that in order to compute ΔS_e as a function of temperature, S_e^{Cu} must be known.

Gold and his co-workers (Ref. 12) have used the Kohler equation to obtain a diffusion thermopower for copper. The thermopower of copper is complicated by the presence of a large negative peak at $T \approx 8$°K. The low-temperature peak is attributed to the presence of trace amounts of iron, which in high-purity, well-annealed copper predominates over other impurities at the very low temperatures (Ref. 12). With respect to diffusion thermopower, the effects of impurities other than iron in high-purity copper cannot be treated with precision using Eq. (3). As an approximation the procedure used by Scott (Ref. 13) is adopted in which the characteristic diffusion thermopower of impurities other than iron is assumed to be given by the Wilson–Sondheimer interpolation formula (Ref. 14 and 15) at low temperatures. Although the interpolation formula gives the wrong sign for the diffusion thermopower of copper, it is treated as a positive quantity and is assumed to give the correct magnitude and temperature dependency (excluding the effects of iron) for the characteristic diffusion thermopower (Ref. 3 and 4). Hence

$$S_e^{Cu} = \frac{(W_e^0 S_e^0 + \Delta W^1 S_e^0 + W_e^{Fe} S_e^{Fe})}{W_e} \qquad (5)$$

where S_e^0 is obtained from the Wilson–Sondheimer formula (Ref. 14 and 15), and S_e^{Fe} and W_e^{Fe} are the diffusion thermopower and electronic thermal resistivity due to the presence of iron in copper, and ΔW^1 is the contribution to electronic thermal resistivity of impurities other than iron, with

$$W_e = W_e^0 + \Delta W \qquad (6)$$

and

$$\Delta W = \Delta W^1 + W_e^{Fe} \qquad (7)$$

The quantity ΔW is obtained from the Wiedemann–Franz law using the residual resistivity of copper from Table 1. The intrinsic electronic thermal resistivity is used to obtain W_e^0 of copper (Ref. 10), and S_e^{Fe} is obtained from Ref. 12. Values for S_e^0 were obtained by first fitting the Wilson–Sondheimer interpolation formula to the data at 320°K (Fig. 1). This procedure, although varying from that used in Ref. 3, is consistent with the assumption of a negligible phonon-drag thermopower in pure copper at this elevated temperature. Assuming a negligible phonon-drag contribution at 8°K, S_e^{Cu} is obtained from the copper data at this temperature (Fig. 1). Hence, using Eq. (5), (6), and (7), W_e^{Fe} is evaluated at this temperature. Assuming the validity of the Wiedemann–Franz law for iron in copper, W_e^{Fe} can then be evaluated as a function of temperature, and S_e^{Cu} can be determined from Eq. (5). The results for S_e^{Cu} are shown in Fig. 1 together with the thermopower of the copper used in this work. The thermopower of copper was obtained by determining ΔS for a copper–lead thermocouple, then using the values for lead determined by Jan, Christian, and Pearson (Ref. 16).

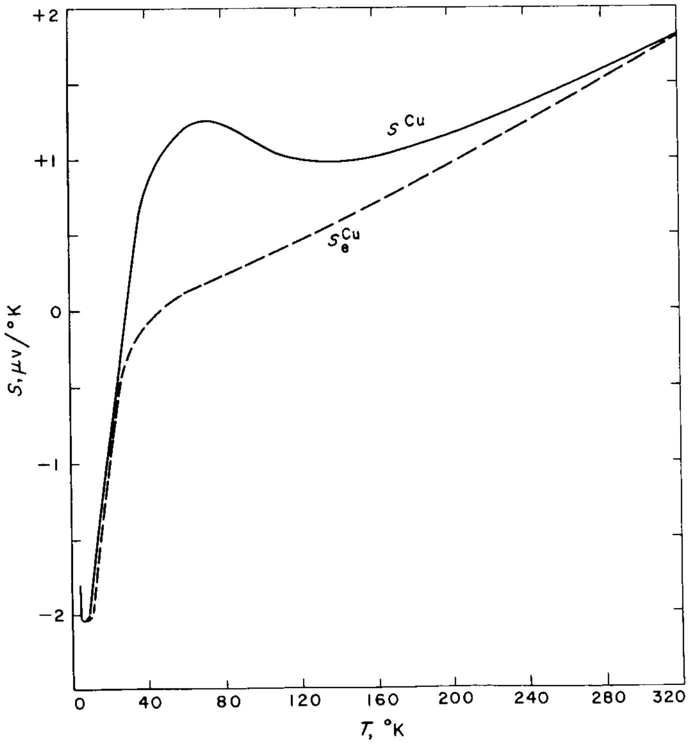

Fig. 1. Thermoelectric power of copper

IV. EXPERIMENTAL RESULTS

Figure 2 shows the assumed phonon-drag component of copper obtained from Fig. 1. The greatest uncertainty in this treatment lies in the low-temperature region; hence, S_g^{Cu} is plotted only for $T > 30°K$. (In the data analyzed in Section V of this Report, the phonon-drag component for $T > 80°K$ is the main concern.)

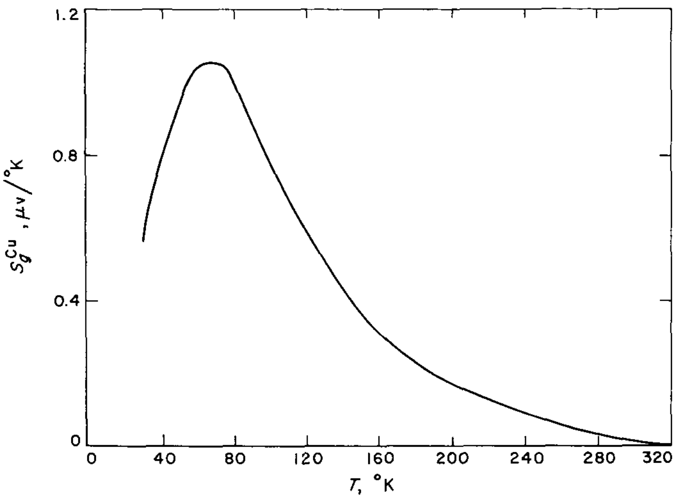

Fig. 2. Phonon-drag thermopower of copper

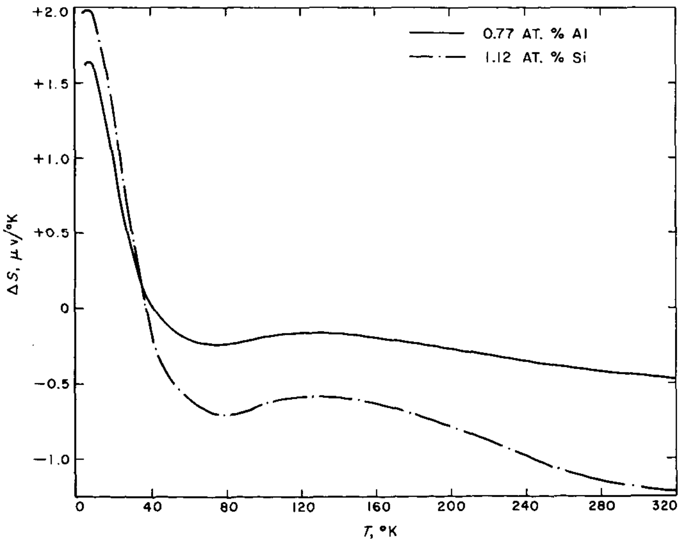

Fig. 3. ΔS, the difference in thermopower between the low-concentration alloys and copper

Use of Eq. (4) in obtaining ΔS_e depends, among other things, on equality of the Fermi energy in both the alloy and pure metal; this is better approximated in the more dilute alloys. In the present case, Eq. (4) is more applicable to the lower-concentration Cu-Al and Cu-Si alloys. The phonon-drag component is thus separated out for the Cu + 0.77 At.% Al and Cu + 1.12 At.% Si alloys using the data at higher concentrations to indicate gross features in the data. The difference ΔS between the absolute thermoelectric power of the pure metal and the alloy is shown in Fig. 3 for the lower concentrations. In Fig. 4, the change in phonon-drag thermopower for these alloys is shown. The current results indicate that phonon-drag attenuation in the Cu-Si alloy is markedly greater than in the Cu-Al alloy. The same holds true in the case of diffusion thermopower as indicated in Fig. 5. This general tendency appears to be continued in the less dilute alloys as indicated by the absolute thermopowers of the higher concentration alloys shown in Fig. 6.

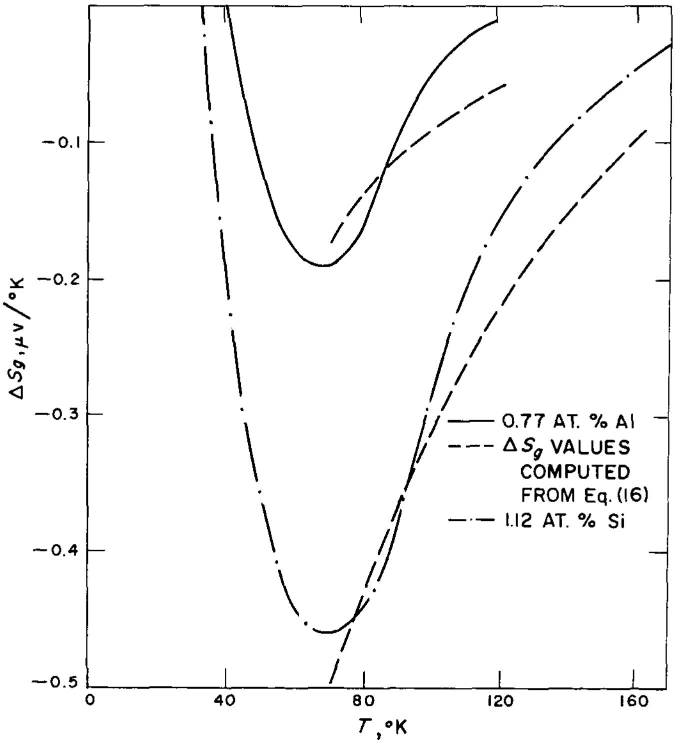

Fig. 4. ΔS_g, the change in phonon-drag thermopower, for the low-concentration alloys

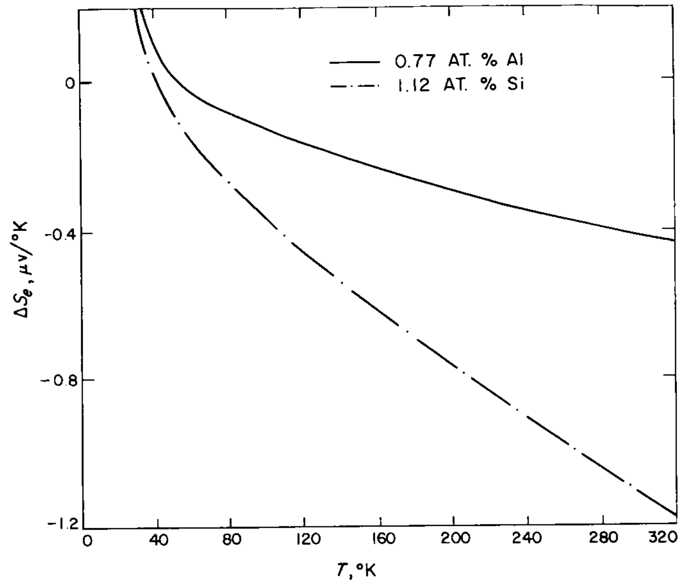

Fig. 5. ΔS_e, **the change in diffusion thermopower for the low-concentration alloys**

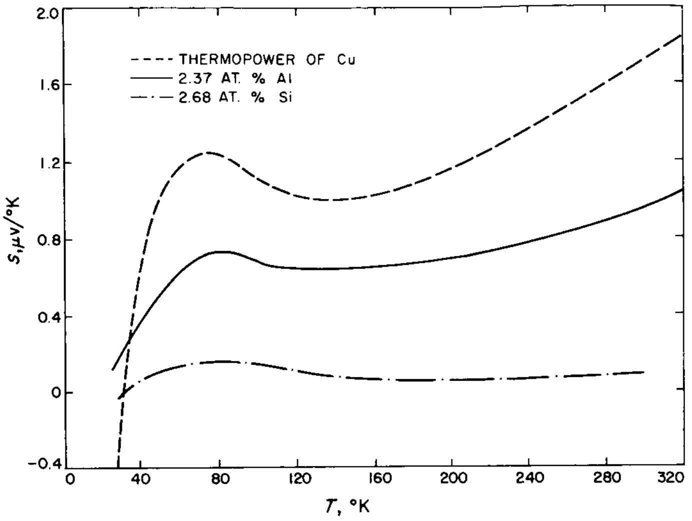

Fig. 6. Absolute thermopowers of the higher-concentration alloys

V. ANALYSIS AND DISCUSSION

Quenching of phonon-drag thermopower in dilute copper alloys has been treated (Ref. 3 and 4) in terms of the theory of Hanna and Sondheimer (Ref. 17). Essentially, the Hanna–Sondheimer theory, by neglecting umklapp processes, yields a relation which has been used as a rough approximation in relating the alloy phonon-drag thermopower to lattice thermal conductivity (Ref. 3 and 4). An alternate method of analysis lies in that developed by Huebener (Ref. 8) for treating phonon-drag attenuation due to lattice vacancies in gold. In the Huebener treatment, both normal and umklapp processes are included in the full expression for the thermopower. A simplification employed by Huebener is the assumption that scattering of phonons by an impurity and all other phonon scattering events are independent of each other (Ref. 8). In this connection, it has been shown that a specific lattice thermal resistivity due to defect scattering can be defined only when this scattering is small (Ref. 18). It is assumed that this is approximately the case in the present low-concentration alloys.

Additional assumptions (Ref. 8) are:

1. Heat transport by phonons is independent of other heat transporting mechanisms;
2. No distinction is made between longitudinal and transverse phonons;
3. The material is elastically isotropic and dispersion in the vibrational spectrum is neglected;
4. A Debye phonon spectrum is assumed;
5. The phonon scattering processes can be represented by frequency- and temperature-dependent relaxation times.

According to Huebener, the change in phonon-drag thermopower ΔS_g of a metal containing defects (for temperatures above the phonon-drag peak) is given by

$$\Delta S_g = -Ae^{\beta/T}\int_0^{\theta/T}\frac{Z^2e^Z\,dZ}{(e^Z-1)^2\,(1+\tau_i/\tau_o)} \tag{8}$$

where θ is the Debye temperature, and τ_o and τ_i are the relaxation times for phonon scattering by the host material and defects, respectively. A and β are constants; $Z = \hbar\omega/kT$, where ω is the phonon frequency; $\hbar$ is Planck's constant divided by 2π; and k is Boltzmann's constant.

The relaxation time τ_o is estimated from the expression obtained by Callaway (Ref. 19) for K_g the lattice thermal conductivity

$$K_g = \frac{k^4T^3}{2\pi^2v_s\hbar^3}\int_0^{\theta/T}\frac{\tau_o(Z)\,Z^4e^Z\,dZ}{(e^Z-1)^2} \tag{9}$$

where v_s is the sound velocity. For $T > 40°\mathrm{K}$, the experimentally determined lattice thermal conductivity of copper is approximated by $K_g = 35T^{-1}$ w cm^{-1} (Ref. 20 and 21). In order to fit the T^{-1} dependency of K_g for pure KCl above the thermal conductivity maximum, Walker and Pohl (Ref. 22) have used the relaxation time

$$\tau_o^{-1} = b\omega^2Te^{-\beta/T} \tag{10}$$

where b is a constant. The relaxation time Eq. (10) also gives a good fit to the lattice thermal conductivity of gold above 30°K (Ref. 8). From Eq. (10) and (11) the following expression is obtained

$$K_g = \frac{k^2e^{\beta/T}}{2\pi^2v_s\hbar b}\int_0^{\theta/T}\frac{Z^2e^Z\,dZ}{(e^Z-1)^2} \tag{11}$$

The integral in Eq. (11) is readily evaluated from tables in Ref. 23. For v_s the velocity of sound in the Debye approximation is used

$$v_s = \frac{k\theta}{\hbar}\left[\frac{\Omega}{6\pi^2}\right]^{1/3} \tag{12}$$

where Ω is the atomic volume. For θ, using the value 310°K obtained from specific heat data (Ref. 24), $v_s = 2.37 \times 10^5$ cm sec^{-1} is obtained. Using $K_g = 35T^{-1}$, it is found from Eq. (11) that $b = 4.19 \times 10^{-18}$ sec deg^{-1}, and $\beta = 43.6°\mathrm{K}$. With the preceding constants in τ_o, Eq. (11) yields a good fit to the lattice thermal conductivity of copper above 40°K.

The relaxation time in Eq. (10) was chosen by Walker and Pohl to give the best fit to their experimental data, no attempt being made at theoretical justification (Ref. 22). Their viewpoint was adopted in this work noting that the same procedure appears to have been followed in the case of gold (Ref. 8). However, it is of interest to explore for possible correlations between β and θ. To do this exploring for the noble metals, it is first noted that the lattice thermal conductivity of silver for $T > 40°\mathrm{K}$ is approximated by $K_g = 14T^{-1}$ w cm^{-1} (Ref. 25). Using $\theta = 220°\mathrm{K}$ (Ref. 24), $\beta = 29°\mathrm{K}$ for silver is obtained from Eq. (11). With the preceding values and using Huebener's values for gold ($\beta = 22°\mathrm{K}$, $\theta = 164.5°\mathrm{K}$), it is seen that $\theta/\beta = 7.5$ for gold, $\theta/\beta = 7.1$ for copper, and $\theta/\beta = 7.6$ for silver. Further comment on this point is dependent on physical justification of Eq. (10). In this case, the numerical value of β was selected using Eq. (11) to give the best fit to the lattice thermal conductivity of copper in the range of 40 to 100°K, the latter temperature representing the maximum value to which K_g is experimentally known (Ref. 20 and 21). Considering, among other things, variations in Debye temperatures (Ref. 24) and the experimental error in K_g, some uncertainty exists with regard to the second significant figure in both β and b. The third significant figure is retained, however, for computational purposes.

The constant A is evaluated from the expression for the phonon-drag thermopower of the pure metal (Ref. 8)

$$S_g^{\mathrm{Cu}} \approx Ae^{\beta/T}\int_0^{\theta/T}\frac{Z^2e^Z\,dZ}{(e^Z-1)^2} \tag{13}$$

Eq. (13) is valid in the region where S_g^{Cu} varies as T^{-1} ($T > 80°\mathrm{K}$). It is obtained from the expression derived by Bailyn (Ref. 26), which includes both umklapp and normal processes for the pure metal and assumes a single normal band of standard form. The constant A contains a factor $(\mathbf{q} + \mathbf{K})/\tau_{ep}$ which is inside the integral, where $\mathbf{q}$ is the phonon wave vector, $\mathbf{K}$ is a reciprocal lattice vector, and τ_{ep} is the relaxation time for electron–phonon collisions. Since this factor is difficult to evaluate, it is treated as an adjustable constant (Ref. 8). From S_g^{Cu} at 90°K (Fig. 2), it is found from Eq. (13) that

$$A = 0.209\mu\mathrm{v}/°\mathrm{K} \tag{14}$$

A pure Rayleigh-type mechanism is assumed for scattering of phonons by the solute atoms; hence

$$\tau_i^{-1} = a\omega^4 \qquad (15)$$

Substituting Eq. (10), (14), and (15) into Eq. (8), it is seen that

$$\Delta S_g = -0.209 e^{\beta/T} \int_0^{\theta/T} \frac{Z^2 e^Z \, dZ}{(e^Z - 1)^2 \left\{ 1 + \dfrac{b\hbar^2 e^{-\beta/T}}{a k^2 Z^2 T} \right\}} \qquad (16)$$

Using the present values of ΔS_g at 90°K, the parameter a is obtained from Eq. (16) by numerical integration. The integration was carried out with an IBM 7090 computer. For Cu + 0.77 At.% Al, the results are

$$a = (0.9 \pm 0.4) \times 10^{-43} \, \text{sec}^3 \qquad (17)$$

For Cu + 1.12 At.% Si, the results are

$$a = (6.5 \pm 2.3) \times 10^{-43} \, \text{sec}^3 \qquad (18)$$

With the preceding values of a and using Eq. (16), ΔS_g is computed as a function of temperature from 70 to 160°K. The results are shown in Fig. 4. The deviations in Eq. (17) and (18) are computed from Eq. (16) using the experimental error in ΔS_g estimated as $\pm 0.05 \, \mu\text{v}/°\text{K}$.

Elastic scattering of lattice waves by point imperfections has been treated by Klemens (Ref. 27). Scattering of phonons by substitutional impurities is attributed to mass difference, changes in the elastic constant of linkages between lattice points, and elastic strain. It was found for these effects that a Rayleigh-type scattering law was obtained (Ref. 27) with

$$a = \frac{3\Omega f}{\pi v_s^3} S^2 \qquad (19)$$

where f is the fraction of solute atoms present in the alloy, and

$$S^2 = S_1^2 + (S_2 + S_3)^2 \qquad (20)$$

with

$$S_1 = \frac{1}{(12)^{1/2}} \left[\frac{\Delta M}{M} \right] \qquad (21)$$

$$S_2 = \frac{1}{(6)^{1/2}} \left[\frac{\Delta F}{F} \right] \qquad (22)$$

$$S_3 = -(2/3)^{1/2} Q\gamma \left[\frac{\Delta R}{R} \right] \qquad (23)$$

where ΔM is the mass difference between the host atom and the solute atom, and M is the average atomic mass for the host material; F is the force constant of an interatomic linkage, and ΔF is the change in force constant of a linkage due to introduction of an impurity atom; R is the nearest neighbor distance, and ΔR is the change in nearest neighbor distance due to a substitutional impurity atom; γ is the Gruneisen constant, and Q is a constant which contains a contribution to the scattering matrix from lattice strain due to other than nearest neighbors.

Using Eq. (21), the effect of mass difference can readily be calculated from Eq. (19). For S_2 and S_3 such physical parameters as bonding of a solute atom and the distortion field are not well known, and only an order-of-magnitude estimate is possible. The calculations are thus restricted to the mass-defect case. Therefore,

$$a_M = \frac{\Omega f}{4\pi v_s^3} \left[\frac{\Delta M}{M} \right]^2 \qquad (24)$$

$$a_M = 1.8 \times 10^{-43} \, \text{sec}^3; \qquad (\text{Cu} + 0.77 \, \text{At.\% Al}) \qquad (25)$$

$$a_M = 2.4 \times 10^{-43} \, \text{sec}^3; \qquad (\text{Cu} + 1.12 \, \text{At.\% Si}) \qquad (26)$$

where a_M is the scattering parameter calculated for mass defect alone.

Comparison of Eq. (17) and (25) indicates that phonon scattering is largely due to mass difference in Cu-Al. On the other hand, for Cu-Si the current results yield a contribution from lattice distortion which is of the same magnitude as the mass-difference term. This is in agreement with the estimated maximum contribution for lattice distortion obtained from the lattice thermal conductivity of Cu-Si alloys evaluated at 90°K (Ref. 5).

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