where

$$F_{k,l}\equiv iY_{k,l}e^{-i(l\varphi+b\sin\varphi)}\sum_{q=-\infty}^{\infty}\frac{J_q(b)e^{i(l+q)\varphi}}{a-l-q}. \quad (50)$$

The quantity $\Gamma_{1,0}^{1,0}$ is proportional to the conductivity $\sigma^0{}_{zz}$ as given in Eq. (6).

V. CONCLUSIONS

The origin and general dispersion characteristics of the high-frequency waves have proved, at least in the case of the ordinary ($\mathbf{J}\|\mathbf{H}$) modes, to be explained quite well by an independent-particle or free-electron model of simple isotropic metals. Residual discrepancies exist, however, and should permit evaluation of several moments of the Landau scattering function which has been used to describe the effects of quasiparticle interactions. In practice, considerable care both in the experiments and their analysis will be required, since only moments V_n, $n\geqslant 2$, produce directly observable results and these higher moments are apparently quite small. We defer this quantitative problem to a later publication.

ACKNOWLEDGMENTS

We have benefited from discussions with numerous colleagues, particularly A. L. McWhorter and D. Hamilton of Lincoln Laboratory (MIT), and P. A. Wolff and S. J. Buchsbaum of Bell Telephone Laboratories.

PHYSICAL REVIEW VOLUME 172, NUMBER 3 15 AUGUST 1968

Lattice Heat Conductivity in Annealed and Quenched Gold-Platinum Alloys*

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(Received 11 March 1968)

The heat conductivity of dilute gold-platinum alloys, containing 0.11 and 1.02 at. % Pt, has been measured between 4.2 and 80°K. The influence of vacancies on the lattice heat conductivity K_g of the alloy with the higher platinum concentration was determined from measurements with specimens which were quenched and subsequently annealed. Above about 30°K, K_g in the annealed alloy Au+1.02 at. % Pt is found to be about 20% smaller than in pure gold. Whereas the lattice heat conductivity is clearly reduced at higher temperatures by quenched-in vacancies, it is practically unaffected by quenching below about 15°K. The phonon-scattering cross section of vacancies is estimated from the data, assuming a Rayleigh-type scattering law. It is found to be in reasonable agreement with the value obtained earlier from the phonon-drag thermopower of quenched and annealed pure gold.

INTRODUCTION

THE lattice heat conductivity and the phonon scattering by lattice defects has been studied in insulators in many experiments.[1] In metals, similar studies are rather complicated because of the dominant conductivity of heat by the conduction electrons. Since, at low temperatures, the lattice component of the thermoelectric power is of the same order of magnitude as or even larger than the electron-diffusion component, it appears to be advantageous to study the scattering of phonons by lattice defects from this property. For instance, it has been suggested from thermoelectric experiments with quenched platinum,[2] that the phonon scattering by vacancies in platinum is characterized by a resonance at low frequencies. On the other hand, it has been pointed out[3–5] that the phonon-drag thermopower may be complicated at low temperatures due to anisotropy of the relaxation times for electron scattering. An apparent anomaly in the phonon-drag thermopower at low temperature might be the result of either one of the mechanisms described.

Recently,[6] we have measured the thermoelectric power of annealed and quenched gold-platinum alloys between 4.2 and 300°K. For an unambiguous interpretation of these experiments it was necessary to study the effect of quenched-in vacancies on the lattice heat conductivity of the alloys. Therefore we measured the thermal conductivity of quenched and annealed gold-platinum alloys between 4.2 and 80°K. Preliminary results of the present investigation were reported earlier.[7]

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

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[1] A. A. Maradudin, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1966), Vol. 18, p. 273.

[2] R. P. Huebener, Phys. Rev. **146**, 490 (1966).

[3] C. van Baarle, Physica **33**, 424 (1967).

[4] M. Bailyn, Phys. Rev. **157**, 480 (1967).

[5] J. S. Dugdale and M. Bailyn, Phys. Rev. **157**, 485 (1967).

[6] R. P. Huebener and C. van Baarle, Phys. Rev. **159**, 564 (1967).

[7] C. van Baarle and R. P. Huebener, Bull. Am. Phys. Soc. **12**, 354 (1967).

TABLE I. Characteristics of the specimens.

Designation	at.%Pt	Treatment	ρ(4.2°K) ($10^{-6}\Omega$ cm)	$\Delta\rho_{vac}$(4.2°K) ($10^{-8}\Omega$ cm)	C_{vac}[a] (at.%)
SA7	1.02	Quenched from 1020°C	0.998	2.6	0.015
SA7*	1.02	Annealed at room temperature 90 h	0.972	···	
SA9	0.11	Quenched from 1000°C	0.154	3.3	0.018
SA9*	0.11	Annealed 60 h at room temperature and 40 h at 43°C	0.121	···	

[a] Calculated with $\Delta\rho_{vac}/c = 1.8\times10^{-6}$ Ω cm/at.% (Ref. 20).

We would expect a resonant scattering of phonons by vacancies to reduce the lattice heat conductivity and the phonon-drag thermoelectric power to approximately the same extent. On the other hand, anisotropic electron scattering would affect the lattice heat conductivity and the phonon-drag thermoelectric power of a quenched specimen quite differently.

The lattice heat conductivity of annealed gold alloys has been determined previously from measurements on gold alloyed with platinum,[8] chromium,[8] and iron.[9] Kemp and co-workers[10,11] have tried to relate the change, due to cold work, in the lattice thermal conductivity at liquid-oxygen temperatures to the increase of the residual electrical resistivity. Assuming that these changes were caused by vacancies introduced in the deformation process, they compared the concentrations of defects calculated from both properties. The discrepancy between the results ranged from factors of 3 to 6. Only mass defect scattering was considered to contribute to the thermal resistance. More recent work has shown that scattering by the distortion of the lattice around the vacancy might outweigh mass-defect scattering by an order of magnitude. Moreover, the kind of lattice defects obtained by cold working is rather unclear.

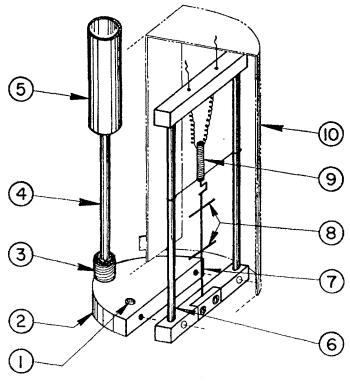

FIG. 1. Specimen arrangement.

[8] J. A. Birch, W. R. G. Kemp, P. G. Klemens, and R. J. Tainsh, Australian J. Phys. **12**, 455 (1959).
[9] G. K. White, S. B. Woods, and M. T. Elford, Phil. Mag. **4**, 688 (1959).
[10] W. R. G. Kemp, P. G. Klemens, A. K. Sreedhar, and G. K. White, Proc. Roy. Soc. (London) **A233**, 480 (1956).
[11] W. R. G. Kemp, P. G. Klemens, R. J. Tainsh, and G. K. White, Acta Met. **5**, 303 (1957).

Therefore, the introduction of vacancies by a rapid quench is preferable.

EXPERIMENTAL PROCEDURE

The specimen material was supplied as polycrystalline alloy wire of 0.010-in. and 0.020-in. diam.[12] The 0.010-in. wire contained 0.11 at.% Pt. It was made from 99.999% pure gold and 99.999% platinum. The 0.020-in. wire contained 1.02 at.% platinum. Its components were specified as 99.9999% gold and 99.999% platinum. Analysis of the material of the wire, however, showed traces of various elements, the total quantity exceeding appreciably the original estimate of impurity concentration.

The specimens were annealed in air for 24 h at 750°C by joule heating, and gradually cooled to room temperature. The wires were quenched from temperatures between 1000 and 1020°C to ice-water temperature within 0.03 sec, corresponding to quench rates of about 3×10^{4}°C/sec. The technique employed has been described before.[13] The concentration of vacancies, introduced in this way, is of the order of 10^{-2} at.%. The specimen characteristics are summarized in Table I.

Within approximately 30 to 40 min after quenching, the specimens were mounted in the cryostat and cooled down to below −40°C. Below this temperature the annealing of vacancies is negligible and the rate of formation of divacancy or vacancy-impurity complexes is probably very low. It is difficult to estimate the configuration of the quenched-in defects finally present in the alloy specimens.

The assembly of the specimen within the vacuum space of the cryostat is shown in Fig. 1. The lower end of the specimen (7) is clamped onto a copper frame (6), which also serves to support the heater (9) by means of the Chromel current leads and a nylon string. The upper end of the specimen is spotwelded onto the gold wire which serves as the heater body.

The frame is connected to the heat sink (5) through a copper platform (2) and a stainless steel tube (4) (0.125-in. diam, 0.010-in. wall thickness). The heat sink is a stainless-steel tube of 0.5-in diam, closed at the

[12] Obtained from Engelhard Industries, Inc., Newark, N. J., and from Cominco American, Inc., Spokane, Wash., respectively.
[13] R. P. Huebener, Phys. Rev. **135**, A1281 (1964).

lower end by a copper block while the upper side is in open communication with the liquid-helium bath. The stainless-steel tube serves as anchoring post to the wires leading into the cryostat from room temperature. Electrical insulation is maintained between the parts of the assembly wherever possible without undoing good thermal contact. Heater (3), wound on the lower end of tube (4), is employed to raise the temperature of the specimen above 4.2°K. The temperature of the platform is measured with a calibrated germanium thermometer, fitting tightly into hole (1). The temperature gradient along the specimen is created by heat flow from heater (9) to the heat sink. This heater is wound from 0.003-in. insulated chromel wire on a gold wire (0.040-in. diam) of 0.5-in. length. The resistance value is about 1000 Ω. This particular construction of the heater, as well as the choice of the thermometers employed, serves to keep the thermal relaxation time of the specimen as short as possible. Because of specimen geometry this time could otherwise become quite long.

The temperature difference is measured between the two points of the specimen, onto which the wires (8) are spotwelded. To avoid contamination the wire pieces (8) are of specimen material. They carry clamping devices which serve to provide a good thermal contact between the specimen and the thermocouples, used as thermometers. We selected a thermocouple made of gold +0.3 at.% iron versus chromel.

The performance of this couple differs somewhat from the gold +0.03 at.% iron couples[14] which are now accepted quite generally in low-temperature research. We will elucidate our choice in the last paragraph of this section. The couple and its performance will be described elsewhere.[15]

The gold-iron alloy wire of 0.001-in. diam and the Chromel wire (0.003-in. diam) are spotwelded to each other. Besides, they are spotwelded onto a gold foil of 0.010-in. thickness, over a length of 0.25 in. to ensure good thermal contact. This foil fits tightly between the clamping devices consisting of a conical copper body and a fitting aluminum ring. The dimensions of this device are very small. The copper body is only 0.20 in. long and has a central circumference of 0.25 in. Moreover, this part was drilled to reduce its heat capacity. All elements—the body, the ring, and the gold foil—have been coated with a thin layer of enamel[16] to provide electrical insulation. Notwithstanding the pressure due to the large thermal contraction of aluminum on cooling, the insulation has always been perfect.

The emf developed by the thermocouple is measured with a Rubicon 6-dial potentiometer; a Keithley 147 serves as null detector. This system has a sensitivity of 2×10^{-9} V. The electrical resistance of the thermocouple is of the order of 20 Ω, mainly located in the 0.003-in. Chromel wires. High resistances are to be preferred to prevent errors in the temperature measurement due to heat leaks. However, because of the high resistance of the couple it is inpractical to construct a reversing switch for eliminating the thermal emf along the extension wires from the arrangement at low temperatures to the potentiometer. Although these emf's are by no means small, they appear to change gradually with the temperature of the specimen. Typically they change by 2 to 3 μV over the temperature range from 4.2 to 80°K. To eliminate the effect of these thermals on the measurement of the temperature differences, the output of the couple is measured twice. First, without a heat current flowing through the sample; the second time with a heat current, while the lower contact point is kept at constant temperature.

Stabilization of the temperature is achieved by means of a feedback control system, which consists of a second thermocouple between the lower contact point and the heat sink, a potentiometer, a null detector, and a controllable power supply. The output of the null detector is fed into the power supply through a circuit to control feedback and bandwidth. In this way a stable control system is obtained which controls the temperature without appreciable overshooting to within 0.005°K.

Chromel wires are spotwelded onto the gold-platinum wires (8) to measure the electrical resistance of the specimen. Thus the geometrical factor is the same for the measurement of the electrical and the thermal conductance, which favors an accurate analysis of the results.

All wires to and from the specimen are thermally anchored on a binding post, situated on platform (2) (not shown in Fig. 1). Frame and platform, including all elements attached to it, are surrounded by a copper radiation shield (10) which is fixed firmly to the platform. The vacuum can, which surrounds the assembly of Fig. 1, is sealed with an indium O ring. During the thermal measurements it is evacuated to a pressure of less than 2.10^{-6} Torr.

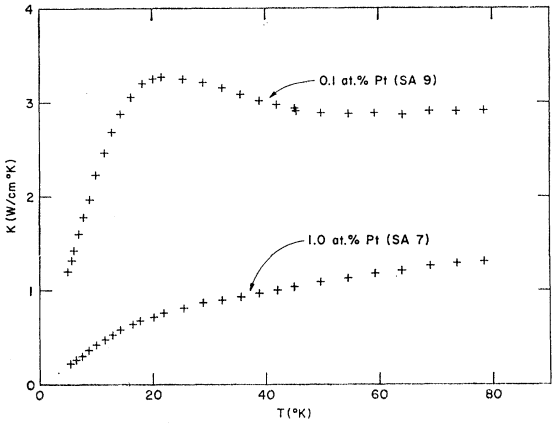

FIG. 2. Total heat conductivity of the annealed alloys containing 0.11 and 1.02 at.%Pt.

[14] R. Berman, J. C. F. Brock, and D. J. Huntley, Cryogenics 4, 223 (1964).
[15] C. van Baarle, R. P. Huebener, and B. Bryson (to be published).
[16] Du Pont VG 8387 Formvar Enamel.

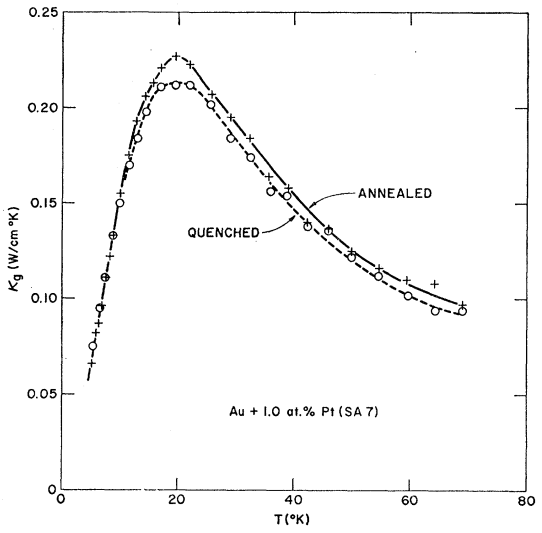

FIG. 3. Lattice heat conductivity of the alloy containing 1.02 at.% Pt in the quenched and the annealed state.

One more remark should be made with respect to the use of thermocouples in an experiment on the thermal conductivity of metals. The electrical insulation between the thermocouple contacts and the specimen introduces unevitably a thermal resistance on these places. It is sometimes suggested[17] that these errors might be eliminated by a two-heater method. This method does indeed allow for the heat leak through the reference leads to the low-temperature heat sink. However, the parallel conduction through the thermocouple branch between the two contact points, in this case the gold alloy wire, causes the measured temperature difference to be smaller by a factor $[1+(W_1+W_2)/W_{\text{gold}}]$ than the actual temperature difference. Here W_1 and W_2 are the thermal resistances of the contacts, W_{gold} is the thermal resistance of the gold alloy wire. The only way to estimate the influence of this error is by repetition of measurements. It is clear that it is reduced by a high value of W_{gold}, which is the reason why we have preferred the high iron concentration.

In preliminary experiments we used another type of clamping to obtain thermal contact between thermocouple and specimen. However, we found that in this way spurious temperature dependences as well as appreciable errors in magnitude were introduced. Through the use of the clamping device described we succeeded to obtain accurately reproducible results for different specimens.

DATA ANALYSIS

The measurements of heat flux and temperature difference provide the total heat conductivity K of the specimen. The separation of K into the electronic contribution K_e and the lattice heat conductivity K_g according to

$$K=K_e+K_g \tag{1}$$

over a wide temperature range can only be accomplished by calculating K_e. The conduction of heat through electrons is limited by impurities and by the electron-phonon interaction. Thus the associated thermal resistance is

$$W_e=K_e^{-1}=W_{\text{imp}}+W_{\text{ph}}. \tag{2}$$

The impurity resistivity is calculated from the electrical resistivity at low temperatures using the Wiedemann-Franz law, while the thermal resistivity due to the phonons is generally taken from measurements on the pure metal. Thus this method assumes the validity of Matthiessen's rule for the thermal resistivity. It is well known[18] that this rule is not a very good approximation at low temperatures. White[19] observed, for relatively pure gold, a marked influence of the purity of the material on the magnitude and the temperature dependence of the phonon resistivity. It is of importance, of course, that the lattice heat conductivity is studied in alloys, where the impurity resistance W_{imp} dominates the electron scattering over a large temperature range.

At high temperatures the total thermal resistivity can be calculated again with confidence using the Wiedemann-Franz law:

$$\rho/W_eT=L_0, \tag{3}$$

where L_0 is the Lorenz number ($L_0=2.45\times10^{-8}V^2/\text{deg}^2$) and ρ is the measured electrical resistivity. It is thus necessary to interpolate by calculation the gap from that temperature, where W_{imp} is no longer dominant, to the temperature range where Eq. (3) can be applied.

For the particular case of the alloy containing 1.02 at.% platinum, we will examplify these arguments numerically at some temperatures between 20 and 80°K. W_{ph} is taken from White's data for the specimen with the lowest heat conductivity. Its electrical resistivity at liquid helium temperatures is 7.6×10^{-8} Ω cm, compared to 9.6×10^{-7} Ω cm for our specimen. Because of the behavior of W_{ph} with increasing impurity concentration, our estimate will give a lower limit of W_{ph}, probably accurate to within a factor of 1.2 to 1.4 at low temperatures. Relative to W_{imp}, W_{ph} amounts to 3.5% at 20°K, 20% at 40°K, and 40% at 60°K. Based on these numbers, we have calculated the lattice heat conductivity, which amounts at the same temperatures to 40, 18, and 10%, respectively, of the electronic conductivity. We expect, because of the matching of K_e at 80°K to the electrical resistivity, the uncertainty in K_g to be largest at about 40°K and to be of the order of 20%.

The additional resistivity due to the quenched-in vacancies does not contribute significantly to W_{imp}, so it does not seriously affect the uncertainty in K_g either. Therefore, it is well possible to observe the small reduc-

[17] R. Berman and D. J. Huntley, Cryogenics 3, 70 (1963).

[18] P. G. Klemens, Australian J. Phys. 7, 64 (1954).

[19] G. K. White, Proc. Phys. Soc. (London) A66, 559 (1953).

tion in K_g as caused by these vacancies by performing two subsequent measurements on the same specimen. The first observation of K is made immediately after the specimens were quenched. The second measurement was made after a room-temperature anneal of the specimen for four to five days without removal of the specimen from the cryostat.

EXPERIMENTAL RESULTS AND DISCUSSION

The total heat conductivity of the annealed alloys containing 0.11 and 1.0 at.% Pt is shown in Fig. 2 as a function of temperature. The heat conductivity for the high-concentration alloy was separated according to Eq. (1) into the electronic and lattice components. The lattice part K_g of this specimen is shown in Fig. 3 for the quenched and the annealed state. Quenching clearly causes a reduction in K_g on the high temperature side of the range, whereas below about 15°K, K_g remains unaffected within the experimental accuracy.

The total conductivity for the quenched and annealed alloy containing 0.11 at.% Pt is shown in Fig. 4 as a function of temperature. Because of the high electronic conductivity in this alloy, the separation of the lattice conductivity from the electronic conductivity could not be done with reasonable accuracy over the whole temperature range. Below 10°K the electronic conductivity was calculated from the electrical resistivity using the Wiedemann-Franz relation. The resulting values for K_e are shown by the dashed straight lines in Fig. 4. Again, at low temperatures, the lattice heat conductivity is apparently not strongly reduced by quenched-in vacancies. The difference between the $K(T)$ curves for the annealed and the quenched alloy can be accounted for within the experimental accuracy by the decrease in K_e due to the additional electron scattering by vacancies. In this alloy the increase of the impurity scattering is of course relatively important, as is clear from the resistivity data in Table I. To check the effectiveness of a room-temperature anneal for the removal of the vacancies, we have also measured the heat conduction of a specimen of the alloy containing 1.0 at.% platinum, carefully annealed for four days at 750°C. The results for this alloy were in good agreement with those obtained on the sample SA 7*, which was quenched and subsequently annealed.

The high-temperature results for both specimens containing 1.0 at.% Pt could be well represented by the relation

$$K_gT=6.3\text{W/cm} \tag{4}$$

at temperatures above 30°K. This value was found by Birch *et al.*[8] for an alloy of gold with 0.7 at.% Pt. In view of the point-defect resistivity of platinum in gold, which amounts to about 0.05 T cm/W at.%, this corresponds to a discrepancy of 10%. Because of the accuracy of the various factors used in the calculation, we regard this agreement as satisfactory.

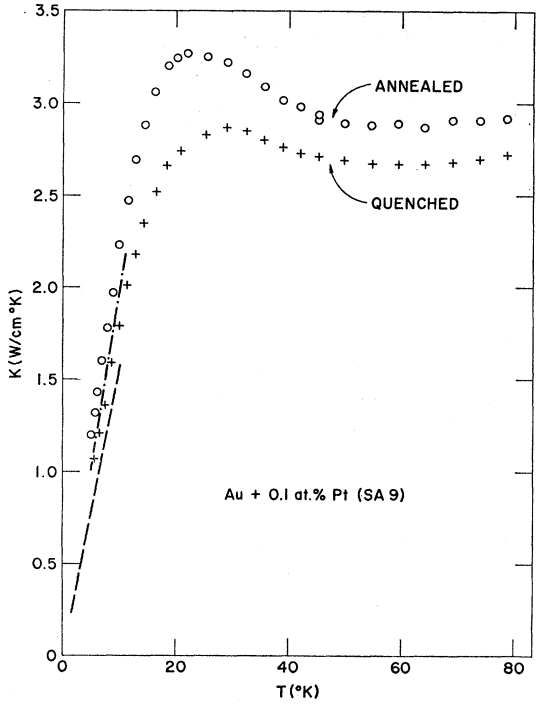

FIG. 4. Total heat conductivity for the alloy containing 0.11 at.% Pt in the quenched and the annealed state.

The lattice heat conductivity passes through a maximum at about 20°K. Well below the temperature of this maximum K_g for these alloys decreases approximately quadratically with temperature. Compared to the 0.7-at.%-Pt alloy, measured by Birch *et al.*,[8] we find a slightly lower conductivity in this temperature range. This results from the reduced electronic mean free path due to the higher platinum concentration.[20]

As mentioned above, the lattice heat conductivity of the alloy containing 1.0 at.% Pt was practically not affected at low temperatures by the lattice vacancies introduced during quenching. We take this to indicate a scattering of phonons by vacancies, which can be described reasonably well by a Rayleigh-type scattering law:

$$1/\tau_{\text{imp}}=a\omega^4. \tag{5}$$

Here τ_{imp} is the relaxation time for scattering by vacancies of phonons of frequency ω. The Rayleigh coefficient a can be obtained from the experimental results in the following way.

In the Debye approximation, the lattice heat conductivity is given by

$$K_g=CT^3\int_0^{\Theta/T} dz\frac{z^4e^z}{(e^z-1)^2}\tau(z). \tag{6}$$

Here C is a constant, Θ is the Debye temperature, and

[20] P. Lindenfeld and W. B. Pennebaker, Phys. Rev. **127**, 1881 (1962).

$z=\hbar\omega/k_BT$. $\tau(z)$ is the effective relaxation time for phonon scattering, determined by the relaxation times τ_i for the different scattering processes according to

$$\tau^{-1}(z)=\sum_i \tau_i^{-1}(z). \tag{7}$$

From Eqs. (6) and (7) it follows that the reduction in K_g due to the admixture of point defects, is given by

$$\Delta K_g=-CT^3\int_0^{\Theta/T} dz\frac{z^4e^z}{(e^z-1)^2}\frac{\tau_0(z)}{1+\tau_{\rm imp}(z)/\tau_0(z)}. \tag{8}$$

We use $\tau_0(z)$ to indicate the relaxation time for phonon scattering in the pure metal. A good fit to the experimentally determined dependence of K_g on T^3 can be obtained by using[13]

$$\tau_0^{-1}(z)=b\omega^2Te^{-\beta/T} \tag{9}$$

with $b=6.96\times10^{-18}$ sec/deg and $\beta=22$°K.

We apply Eqs. (8) and (9) to the case of pure gold and the reduction caused by the platinum admixture of the alloy. The reduction ΔK_g, relative to K_g for pure gold, is only 20%. From this we obtain

$$\frac{\Delta K_g}{K_g}=-\frac{1}{b}e^{\beta/T}T\left(\frac{k_B}{\hbar}\right)^2\frac{J_4^*(\Theta/T)}{J_2(\Theta/T)}a. \tag{10}$$

The transport integrals $J_n(\Theta/T)$ are given by

$$J_4^*(x)=\int_0^x dz\frac{z^4e^z}{(e^z-1)^2}\frac{1}{\alpha z^2+1},$$

with

$$\alpha=\frac{a}{b}\left(\frac{k_B}{\hbar}\right)^2Te^{-\beta/T} \tag{11}$$

and

$$J_2(x)=\int_0^x dz\frac{z^2e^z}{(e^z-1)^2}.$$

The contribution from αz^2 in $J_4^*(x)$ is relatively small, because $\tau_{\rm imp}(z)/\tau_0(z)$ is small. With $\Delta K_g/K_g=-0.2$, we calculate, with Eq. (10),

$$a/c=(0.53\pm0.05)\times10^{-42}\ \text{sec}^3/\text{at.}\% \tag{12}$$

for the scattering parameter of platinum ions in gold.

As shown in Fig. 3, the introduction of vacancies into sample 7 reduced the lattice heat conductivity of the alloy by about 5%. Therefore, the scattering associated with the vacancies corresponds to a Rayleigh coefficient of about one-quarter of the value given in Eq. (10). With the quenched-in electrical resistivity $\Delta\rho_{\rm vac}$ of 2.6×10^{-8} Ω cm and the resistivity-per-vacancy concentration[21]

$$\Delta\rho_{\rm vac}/c=1.8\times10^{-6}\ \Omega\ \text{cm/at.}\%, \tag{13}$$

we obtain for the Rayleigh coefficient a^*, describing the phonon scattering by vacancies,

$$a^*/c=(8\pm2)\times10^{-42}\ \text{sec}^3/\text{at.}\%. \tag{14}$$

This value is in reasonable agreement with the value

$$a^*/c=(20\pm6)\times10^{-42}\ \text{sec}^3/\text{at.}\%$$

obtained from the change due to quenched-in vacancies[13] in the phonon-drag thermopower of pure gold. It is appreciably larger than the value of the scattering parameter, calculated for mass-difference scattering alone.[13] This is not too surprising, as from experiments on phonon scattering by F centers in alkali halides[1] it is known that phonons are predominantly scattered by the strain field associated with the vacant lattice site.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge many discussions with R. E. Govednik regarding the experimental arrangements. Some parts of the experiments were carried out by D. S. Jordan and by B. Bryson. One of us (C. van Baarle) expresses his gratitude to the members of the Solid State Science Division at Argonne National Laboratory for their kind hospitality.

[21] R. P. Huebener and C. G. Homan, Phys. Rev. **129**, 1162 (1963).