PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON CRYSTAL LATTICE DEFECTS, 1962, CONFERENCE
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 18, SUPPLEMENT II, 1963

Phonon Scattering by Dislocations

S. ISHIOKA AND H. SUZUKI
Japan Atomic Energy Research Institute
Tokai-mura, Ibaraki-ken, Japan

Thermal conductivities of NaCl crystals containing various numbers of dislocations were measured at temperatures between 4° and 13°K. The observed thermal conductivities are expressed by $K=1.4\times10^5T^2/N$ watts cm^{-1} deg^{-1}, where N is the dislocation density. The phonon scattering probability by dislocations is about 1000 times larger than the theory by Klemens as observed by Sproull *et al.* in LiF. A much larger scattering cross section is expected in vibrating dislocation, and a quantum mechanical calculation was carried out assuming a simplified interaction between phonons and a dislocation. Although the scattering cross section increased considerably, it was not enough to explain the experiments, especially for high frequency phonons. The discrepancy between the theory and the experiments seems to be due to the oversimplified interaction Hamiltonian used in the calculation.

1. Introduction

Lattice imperfections scatter phonons and have an important effect upon the thermal conductivity at low temperatures. The theory of the effect has been given by Klemens[1]. According to his theory the lattice thermal conductivity of the crystal containing dislocations, K, should be proportional to T^2 in the He temperature region. This temperature dependence of thermal conductivity has been observed in cold worked Cu-alloys[2] and LiF crystals[3]. The observed phonon scattering width of dislocations in LiF, however, was to be too large by roughly a factor 1500 compared with the theory. Experimental results in Cu-alloys did not show such a large discrepancy; the factor seems to be less than ten. In this paper the measurements of the thermal conductivities in deformed NaCl crystals are reported and it is shown that the similar discrepancy with LiF exists in NaCl. A calculation on the scattering by the vibrating dislocation model is also discussed in order to improve the theory.

2. The Experimental Method and the Results

The thermal conductivities of cold-worked NaCl crystals were measured at He temperature region. The specimens were cleaved from a NaCl crystal made by Horiba Co. The size of specimens was $8\times8\times50$ mm. These were annealed in vacuum sealed glass tube for 50 hours at 480°C, and compressed by 2.0, 3.0, 6.2 and 7.0% in length, respectively. The dislocation density was determined by counting the etch pits, which were revealed by Schmitten and Haasen's method after the conductivity measurements. A photograph of an etched surface of 6.2% compressed specimen is shown in Fig. 1. The cryostat used in the measurements is shown schematicaly in Fig. 2. The temperature gradient was measured by means of two carbon resistors. The thermal contact between specimen and the the thermometer is similar to that used by Slack in his measurement on KCl[4].

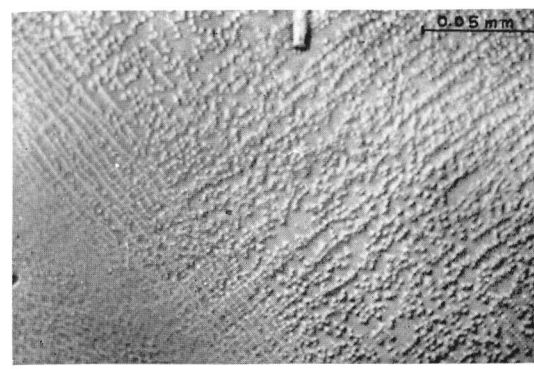

Fig. 1. Freshly cleaved surface of 6.2% compressed NaCl etched to reveal dislocations.

Results of the measurement are shown in Fig. 3. The dotted curve in this figure is the conductivity of very pure crystal measured by Klein[5]. The ultraviolet absorption of the crystal used in this investigation is shown in Fig. 4. An absorption at about 185 mμ, which is referred to as the OH-band, was considerably weak compared with that of Harshaw's one[5]. This may be the reason why the conductivity

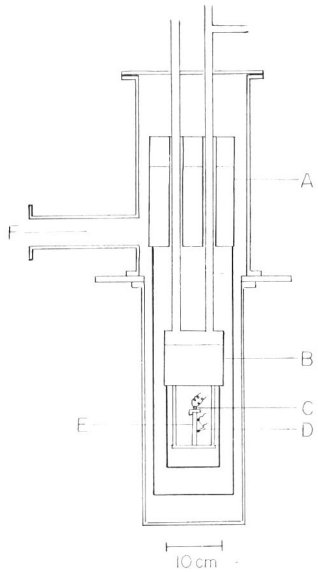

Fig. 2. Schematic drawing of the cryostat. A: liquid N_2 chamber, B: liquid He chamber, C: heater, D: carbon resistor, E: specimen, F: pipe to pumps.

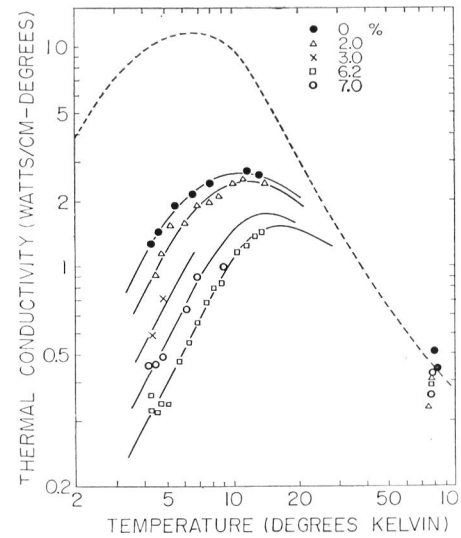

Fig. 4. Thermal conductivity of NaCl crystal as a function of absolute temperature. Dislocation densities of specimens compressed 0, 2.0, 6.2 and 7.0% are 1.2, 4.5, 9.3 and 8.5×10^6 cm^{-2}, respectively.

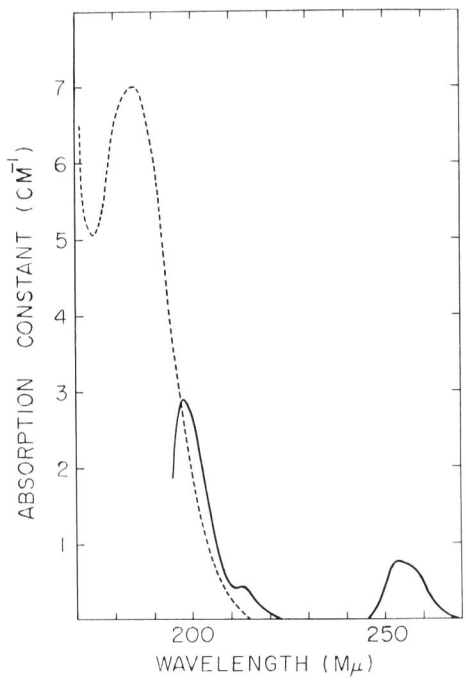

Fig. 3. Ultraviolet optical absorption of an as-cleaved crystal. The dotted curve shows absorption of Harshaws crystal measured by Klein[5].

of as annealed specimen is larger than that of Harshaw's. Other absorption peaks by T1 were observed at 198 mμ and 253 mμ, whose absorption constant were 3.0 and 1.0, respectively. The phonon scattering, however, is not sensitive to T1. The observed conductivity is proportional to T^2 as expected from the theory of Klemens. From these measurements we obtain the relation

$$K=1.4\times10^5T^2/N \qquad (1)$$

where N cm^{-2} is the dislocation density, while the theory by Klemens gave

$$K=4.6\times10^8T^2/N. \qquad (2)$$

Comparing (1) with (2), it is concluded that the phonon scattering width of dislocations in NaCl is larger than the theoretical value by the factor about 1000 as in the case of LiF.

3. The Vibrating Dislocation Model

Carruthers[6] calculated the scattering width of the dislocation more directly than Klemens and his results agreed with Klemens' one. It seems, therefore, that the exact value of the scattering width of the strain field around the dislocation is not so far from their results. In the case of phonon scattering by the isotope, the Callaway model removed the discrepancy between the experiment and the theory[7]. The essence of the Callaway model is that the

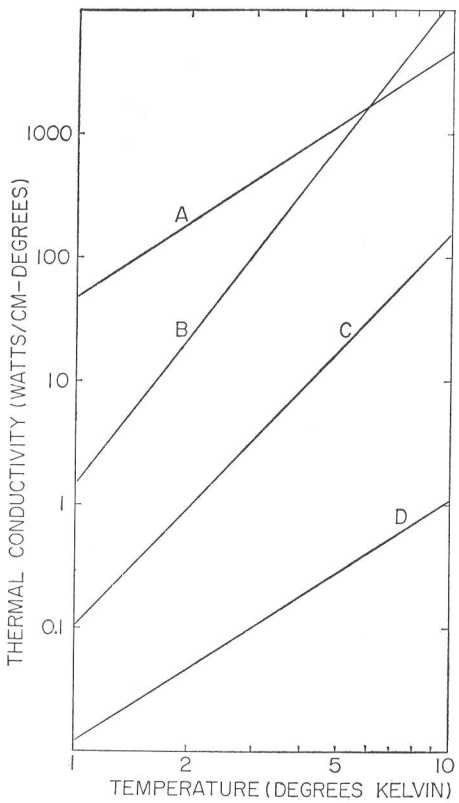

Fig. 5. Comparison of the experimental and theoretical values for the phonon scattering by dislocations in NaCl. A: Klemens theory, B: this calculation, C: Nabarro-Granato theory, D: experiment. Theoretical values are calculated assuming that phonons are scattered only by dislocations and that the dislocation density is $10^7\,\mathrm{cm}^{-2}$. Experimental values are that of specimen reduced by 6.2% in length which exhibited $9.3\times10^6\,\mathrm{cm}^{-2}$ etch pits.

contribution of 3-phonon normal process was added to the thermal resistivity. In the case of the dislocation, however, N-process does not contribute very much to the resistivity and the Callaway's method cannot remove the discrepancy. Other possible processes which would enlarge the thermal resistivity due to dislocations, for example, the coherent addition of scattering amplitudes from neighbouring dislocations or the enhancement of phonon-phonon scattering by strain field, are insufficient to explain this discrepancy. The scattering due to the strain field, therefore, does not seem to be the dominant mechanism of the thermal resistivity by dislocations in ionic crystals.

Klemens and also Carruthers have assumed that the dislocations do not move. Dislocations, however, can vibrate, and absorb and emit phonons. In other words, there are localized lattice vibration modes around the dislocation which are excited by phonons and decay by emitting phonons. The probability of this kind of phonon scattering may be sufficiently large to account for the observation, if there are vibrational states of the dislocation with low frequencies.

Consider a part of dislocation with Burgers vector $\boldsymbol{b}(b_x,\ 0,\ b_z)$ which lies along the Z-axis between $Z=-L/2$ and $Z=L/2$, and vibrates in the valley of Peierls-Nabarro potential. The dislocation can be regarded as the string which has a mass M per unit length, a line tension W, and is subjected to a Peierls-potential U. For the small vibration U is given by $Pf^2/2$ where P is a constant, and $(f(z),\ 0,\ z)$ represents the position of the dislocation. Lagrangian for small vibration is then

$$\mathscr{L}=\int_{-L/2}^{L/2}\frac{1}{2}\left\{M\left(\frac{\partial f}{\partial t}\right)^2-Pf^2-W\left(\frac{\partial f}{\partial z}\right)^2\right\}dz. \quad (3)$$

This is a one-dimensional scalar field equation. From this equation the energy of the vibrating dislocation is obtained in customary way:

$$H=\sum_n E_n\left(a_n^*a_n+\frac{1}{2}\right) \quad (4)$$

$$E_n=\hbar v_s\sqrt{m^2+p_n^2} \quad (5)$$

$$p_n=\frac{n\pi}{L}\quad (n=\pm1,\ \pm2,\cdots) \quad (6)$$

where a^*, a are a creation and an annihilation operator of the quantum of the vibrating dislocation. m is the mass of the quantum which is given by $(P/W)^{1/2}$.

Next, let us consider the interaction between the vibrating dislocation and phonons. The force on the part $d\boldsymbol{x}$ of the dislocation due to the stress field $\boldsymbol{\sigma}$ is given by

$$\boldsymbol{F}=(\boldsymbol{b}\boldsymbol{\sigma})\times d\boldsymbol{x}\ .$$

The interaction energy may be denoted as the work done by the force

$$H_{\mathrm{int}}=\int_{-L/2}^{L/2}(\sigma_{yz}b_z+\sigma_{yx}b_x)f\,dz \quad (7)$$

where $\boldsymbol{\sigma}$ is the stress tensor produced by phonons.

The interaction between the dislocation and the phonon is analogous to that between an atom and a photon, and the scattering probability can be calculated in the similar way with that in the case of photon. Phonons are

absorbed and re-emitted by the dislocation if the energy of a vibrating dislocation is the same as the energy of phonons. It is, therefore, assumed that phonons with different energies are not scattered, namely, the resonance scattering with very sharp peaks. According to this assumption the scattering width is expressed by the δ-function. Taking the average over dislocation length L, where probability distribution is assumed to be proportional to $\exp(-L/L_0)$, the scattering width is given as follows

$$\sigma_\lambda(E,q_z)=\frac{\mu^2b^2(n_P+1)}{2\hbar L_0^2W\rho v_\lambda}\frac{EB}{P^2}\times\left[\frac{1}{(P-q_z)^2}\left\{\frac{1}{\exp\left(\frac{\pi}{L_0P}\right)-1}-\frac{\exp\left(\frac{\pi}{L_0P}\right)\cos\left(\frac{P-q_z}{P}\pi\right)-1}{\exp\left(\frac{2\pi}{L_0P}\right)-2\exp\left(\frac{\pi}{L_0P}\right)\cos\left(\frac{P-q_z}{P}\pi\right)+1}\right\}+\frac{1}{(P+q_z)^2}\left\{\frac{1}{\exp\left(\frac{\pi}{L_0P}\right)-1}-\frac{\exp\left(\frac{\pi}{L_0P}\right)\cos\left(\frac{P+q_z}{P}\pi\right)-1}{\exp\left(\frac{2\pi}{L_0P}\right)-2\exp\left(\frac{\pi}{L_0P}\right)\cos\left(\frac{P+q_z}{P}\pi\right)+1}\right\}\right] \tag{8}$$

where the symbols have the following meanings:

L_0: mean length of the dislocation

$P(E)=\left(\frac{E}{\hbar v_s}-m^2\right)^{1/2}$: wave number of vibration of the dislocation

B: a function of direction of phonon propagation, polarization and Burgers vector (this value is about 0.3)

v_λ: velocity of sound (λ is the polarization index)

$n_P=\frac{1}{\exp(E_P/KT)-1}$: number of quanta of the vibrating dislocation with wave number P

q_z: Z-component of wave vector of phonon

ρ: density of the crystal

μ: shear modulus.

Fig. 5 shows the result of the calculation, where m and L_0 are assumed to be 0 and 1000 Å respectively. The result is not very sensitive to these values.

4. Discussion

The calculation described in the previous section is not sufficient to interpret the large effect of dislocations on the thermal resistivity of ionic crystals. This is due to the incompleteness of the interaction Hamiltonian between phonons and dislocations. The interaction (7) is the static one, which is correct only for the quasi-static motion of dislocation. This corresponds to the approximation in which the interaction between photons and a charged particle is expressed only in terms of scalar potential. It is, however, well known that the interaction can be expressed by the aid of vector potential. A more correct interaction than (7), which may be called as dynamical interaction is, therefore, given by a corresponding function to the vector potential in electromagnetism. Although the calculation using the revised interaction is only at the preliminary state, the scattering probability increases significantly compared to the static interaction theory.

The scattering of sound wave by a vibrating dislocation was already discussed by Nabarro[8] for a different purpose, and his result was applied to calculate the thermal conductivity by Granato[9]. Nabarro's calculation was a classical version of the calculation in the previous section. The difference between the classical and the quantum mechanical calculations is significant in crystals with large Peierls force. If the Peierls force is sufficiently large, phonons are scattered as in the case of resonance scattering. The situation becomes very similar to the scattering of phonons in KCl by NO_2 which was measured and compared with the theory by Pohl[10]. The thermal conductivity in that case reveals the similar temperature dependence to that of crystals containing dislocations. The temperature dependence of the thermal conductivity may, therefore, be obtained correctly by the quantum mechanical theory taking into account the Peierls force.

The thermal conductivities of cold-worked Cu-alloys showed fairly well agreement with the strain field scattering theory. In this case, the problem is complicated by conduction

electrons. It must be remembered that the electron scattering due to a dislocation deduced from electrical conductivity measurements is much larger than that predicted by the theory. It has been believed that an electron is strongly reflected by the stacking fault between two half dislocations, but the electrical resistivity due to dislocations in Al is not especially small. This fact suggests that a dislocation itself interacts more strongly than theory predicted. We may expect larger scattering probability due to an inelastic scattering, in which electrons are scattered by giving energy or taking energy from the vibrating dislocation. A preliminary calculation on this problem indicates that a dislocation interacts with electrons much more frequently than phonons. Since the dislocation at the vibrational states in thermal equilibrium does not interact with phonons frequently, the chance of interaction between phonons and the vibrating dislocation in a metal is considerably less than insulator. This may be the reason why the strain field scattering theory can explain experimental results in Cu-alloys. However, the above mentioned estimations on the scattering of phonons and electrons due to the vibrating dislocation are only preliminary ones and more detailed calculations are required.

References

1 P. G. Klemens: Proc. Roy. Soc. **A208** (1951) 1113.
2 W. R. G. Kemp, P. G. Klemens and R. J. Tainsh: Phil. Mag. **4** (1959) 845; J. N. Lomer and H. M. Rosenberg: Phil. Mag. **4** (1959) 467.
3 R. L. Sproull, M. Moss and H. Weinstock: J. Appl. Phys. **30** (1959) 334.
4 G. A. Slack: Phys. Rev. **105** (1957) 832.
5 M. V. Klein: Phys. Rev. **122** (1961) 1393.
6 P. Carruthers: Rev. Mod. Phys. **33** (1961) 92.
7 J. Callaway: Phys. Rev. **113** (1959) 1046; J. Callaway: Phys. Rev. **122** (1961) 787.
8 F. R. N. Nabarro: Proc. Roy. Soc. **A209** (1951) 278.
9 A. Granato: Phys. Rev. **111** (1958) 740.
10 R. O. Pohl: Phys. Rev. Let. **8** (1962) 481.

DISCUSSION

Klemens, P. G.: The mechanism suggested here finds support from Dr. Sproull's annealing curve; his curve shows that the resistance disappear when the dislocations line-up in walls. Contrary to Dr. Sproull's remark, the line-up would not quench the ordinary phonon scattering, but the dislocations are restrained from vibrating when lined up, and if the resistance arises from dislocation vibration, the observed behavior follows naturally.

Queisser, H.: Hubner (Shockley Transistor) has recently performed measurements on silicon, studying phonon-scattering at dislocations by means of the "transmitted phonon-drag". From the results he concludes that a "vibrating string"-mechanism accounts for the scattering. The scattering cross-section increases with phonon wavelength. Pinning of the dislocations by Cu-precipitation reduces the scattering.

Brandon, D. G.: Have you any idea of the amplitude of vibration that is involved?

Ishioka, S.: The theory demands that the dislocation should stay in one Peierls valley, but it might be extended to amplitudes of vibration greater than the lattice spacing.

Gilman, J. J.: The discrepancy between the theory and experimental results on the thermal resistance of dislocations may be accounted for by the large concentrations of edge dislocation dipoles that are produced during plastic deformation. Evidence of these in NaCl has been given by Whitworth and Davidge (Phil. Mag. (1961)), and the concentration (which would be proportional to the dislocation monopole density) is expected to be roughly 10^3 times the monopole density in order to account for strain-hardening (Gilman: J. Appl. Phys. (August 1962)).

Since dipoles anneal out before monopoles, this interpretation accounts for the more rapid annealing of thermal resistance compared with dislocation density as reported by Sproull and Pohl in the previous paper.

Edge dislocation dipoles can scatter phonons *via* various modes. It is not clear at present which mode will be dominant; and the dominant mode will depend on the

phonon wavelength as well as the glide plane spacing of the dipole. Considered as a line (phonon wavelength comparable to glide plane spacing), a dipole is both a center of lattice rotation and of mechanical polarizability (low shear modulus). For shorter phonons it is two separate dislocations with modified strain fields. Also, it may have some point character at its ends; and at any orientation it junctions along its length.

PROCEEDINGS OF THE INTERNATIONAL CONFERENCE ON CRYSTAL LATTICE DEFECTS, 1962, CONFERENCE
JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 18, SUPPLEMENT II, 1963

An Experimental Study on Electrical Resistivities of Vacancy, Dislocation and Stacking Fault in Aluminum

Sho YOSHIDA, Takao KINO, Michio KIRITANI, Satoru KABEMOTO
Hiroshi MAETA AND Yoshiharu SHIMOMURA

Laboratory of Crystal Physics, Hiroshima University, Japan

The change in electrical resistivity of super pure aluminum due to the lattice defects introduced by quenching and by deformation is correlated with the kinds and concentrations of defects obtained by transmission electron microscopy. The resistivity of an ordinary dislocation does not come from the stacking fault but mainly from the dislocation itself. The specific resistivities of vacancy and dislocation and the resistivity of stacking fault are found to be $10^{-26}\Omega$ cm, $10^{-19}\Omega$ cm and $10^{-13}\beta\Omega$ cm^2 respectively, for stacking fault density β cm^{-1}.

§ 1. Introduction

There has been considerable interest in the discrepancy between the calculated and experimental values for the increment in electrical resistivity due to dislocations in deformed metals. Seeger[1] and Howie[2] have explained this discrepancy by the resistance due to the stacking faults separating the partial dislocations.

The decrement of electrical resistivity by the disappearance of tetrahedra containing the stacking fault in quenched gold foil was measured by Cotterill[3] and this resistivity decrement was interpreted as the annealing out of the stacking fault.

Recently it has been found that almost all dislocation loops in quenched pure aluminum contain a stacking fault in spite of its high stacking fault energy[4]. Using this fact, the resistivity of the stacking fault would be separated from that of dislocation itself. With the aim to determine which is mainly effective to the dislocation resistivity, dislocation itself or stacking fault, the electrical resistance measurements have been carried out on quenched and on deformed aluminum, together with the direct observations of the defects by transmission electron microscopy.

§ 2. Deformation Experiments

Single crystals of 99.998% pure aluminum, which were prepared by Fujiwara's stress-annealing method, were used for the deformation experiments. Those single crystals had their lengthwise axis in the direction shown in Fig. 1 and are considered to be deformed predominantly by the single glide in the early stage of elongation. The specimens for resistivity measurement were single crystal wires of 1.6 mm in diameter, on which four pieces of fine wires of the same material were spot-welded, two for potential leads and two for current leads. The distance between

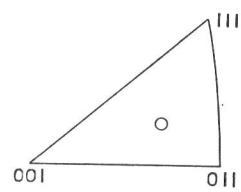

Fig. 1. Crystallographic orientation of lengthwise direction of crystals for deformation experiment.