Electrical Resistivity of Dislocations in Metals

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FUSION TECHNOLOGY INSTITUTE

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INTRODUCTION

Efforts to determine the contribution of dislocations to the electrical resistivity of metals have been rigorously pursued for the past thirty years. The electrical resistivity of dislocations is of considerable interest on purely theoretical grounds, but perhaps more importantly it is of significance in applied situations. One example of its practical use is the determination by resistivity methods of the magnitude of neutron radiation damage to metals. Energetic neutrons such as those found in nuclear fission or proposed nuclear fusion reactors create atomic displacements which may aggregate into loops of dislocation lines. This radiation damage affects the mechanical and electrical properties of the metal, and the magnitude of the effect may be conveniently measured by resistivity techniques.

This paper will outline the present theory of the electrical resistivity of dislocations. Particular attention will be given to the theory developed by Brown, which is in satisfactory agreement with experimental results. A brief summary of early theoretical attempts to calculate dislocation electrical resistivity will be presented first. This will be followed by some comments regarding experimental methods and a review of the Brown model.

INITIAL THEORETICAL MODELS

The first attempt to calculate the electrical resistivity due to a dislocation line was made by Koehler[1] in 1949 using first order perturbation theory. He modeled the dislocation as having the potential of a rigid ion with a screened Coulomb field. His calculated resistivities were of a reasonable order of magnitude, but the applicability of perturbation theory was questioned for this case where displacements caused by the dislocation can be as large as half a lattice spacing. Also, he had to introduce a cut which

joined pairs of dislocations in order to make the displacement single-valued. Subsequent approaches introduced by various researchers allowed the screening electrons around each ion to redistribute so as to keep the Fermi level constant. The electrostatic potential then varies with location in the crystal, and scattering is proportional to the lattice dilatation. This approach eliminated the discontinuity in the scattering potential that was present in Koehler's model. Hunter and Nabarro[2] modified the theory by using a deformation potential that took account of both the interaction of conduction electrons with the ion cores and their interaction with the displaced electronic charge. They found that each conduction electron interacts with both the distorted lattice potential and with the electrostatic field of the space charge, with a large cancellation between these effects. This approach is applicable only when the strain varies slowly, and is therefore not appropriate near the core of the dislocation. Calculation of the dislocation resistivity using this model yields results smaller than experimentally observed values by a factor of 50 for the case of copper. Harrison[3] considered the effect of dilatation in the dislocation core by using a simple model where the core is a hollow cylinder which contains the volume increase associated with a dislocation, corresponding to one vacancy per atomic plane. Using the Born approximation, he obtained a resistivity which was about a factor of 5 below the observed value for copper. This approach is questionable since the second-order long-range elastic strain field appears to be responsible for most of the volume change associated with dislocations, so this volume change should not all be assigned to the core region. More detailed reviews of the above theories may be found in the literature.[4,5]

The discrepancy between the early theories and experimental values of the dislocation resistivity led Broom[6] to suggest that stacking faults may be responsible for an appreciable amount of the measured resistivity in deformed metals. Stacking faults are two-dimensional imperfections in a crystal arising from an alteration in the stacking sequence of the atomic planes (see, e.g. Ref. 5). They are formed in FCC metals when a dislocation dissociates into the more energetically favorable configuration of two partial dislocations. The width of the stacking fault ribbon depends on the stacking fault energy of the metal in question, which generally is not well known. It is assumed that the stacking fault ribbon reflects a certain portion of the conduction electrons incident on it, but the reflection coefficient R is difficult to calculate and various theories predict values of R ranging over several orders magnitude. The value of R, which varies with angle, depends on the form of the Fermi surface and the width of the energy gap there. Only electrons on the Fermi surface are expected to be strongly reflected. The situation prior to 1960 has been reviewed by Ziman[5] and Howie.[7] The resistivity of a stacking fault is given by

$$\rho = \frac{A}{vol} \frac{\hbar}{e^2} (2\pi)^3 \Big/ \int_{\text{Fermi surface}} \frac{|\cos \theta|}{R} \, d\omega_k \qquad (1)$$

where A is the total stacking fault area.

The first reliable estimate of R was made by Howie,[7] who considered conduction electron scattering as a diffraction mechanism and assumed the two-beam approximation (transmitted beam plus diffracted beam from one set of crystal planes) was valid. Using the free electron approximation, he concluded that the scattering of conduction electrons by the stacking fault was

due to the phase change of electrons which are diffracted from non-parallel crystal planes that do not contain a displacement vector of the fault. Howie calculated that the reflection coefficient R was on the order of 0.08-0.30, and that electrons incident normal to the stacking fault are not scattered. This approach was criticized by Seeger and Statz,[8] who claimed that the approximation of free electrons is impermissible and they also questioned the use of the two-beam approximation for the relatively low energy conduction electrons. Starting from the linearized Boltzmann equation and using perturbation theory, they deduced a value of R on the order of 0.5. They also determined that electrons incident normal to the stacking fault were completely reflected. The results of their calculation depend critically on detailed assumptions and it is not clear whether their or Howie's approach is more correct, although some experimental evidence tends to support Howie's result. Even with these large values of R, unreasonably wide stacking fault ribbons are necessary in order to make the calculations agree with the observed dislocation resistivity.

Experimental measurement of the stacking fault resistivity was first performed in 1961 by Cotterill,[9] who rapidly quenched gold foils so as to produce excess vacancies which condense into stacking fault tetrahedra. This and subsequent work performed on metals with stacking fault ribbons formed from dislocation dissociation indicated that in general stacking fault ribbons have an insignificant effect on the dislocation resistivity. Therefore, the situation in the early 1960's still showed a discrepancy between theory and experiment on the order of a factor of 50.

Accurate experimental measurements of the dislocation resistivity are hampered by several difficulties. First, controlled annealing procedures,

which are designed to not eliminate too many dislocations, must be followed in order to separate out the resistivity effects due to other imperfections (in particular vacancies). The resistivity due to point defects created by cold-working always exceeds that due to the created dislocations. Therefore, adequate time must be given to allow the point defects to anneal out of the crystal during the heat treatment. Second, it is difficult to obtain a reliable estimate of the number of dislocations which contribute to the observed resistivity change. Direct observation by transmission electron microscopy (TEM) methods is the best way to determine number densities. Also, aggregates of point defects which are too small to be observed by TEM may contribute to the measured resistivity. Therefore, experimental results are generally only accurate to within a factor of two. With proper experimental precautions, the above-mentioned error may be reduced to about ten percent.

The first dislocation resistivity model which was in reasonable agreement with experimental data was developed by Basinski et al.[10] They proposed a semi-empirical theory in which the resistivity was assumed to be proportional to the mean-square displacement of the ions from their unperturbed positions. This reasoning was based on experimental evidence which indicated that the ratio of ideal resistivity to thermal energy is roughly constant over a wide range of temperatures (and therefore a wide range of phonon wavelengths). Thus they assumed that lattice deformation around a dislocation is similar to the deformation due to thermal vibrations. For a crystal with atomic volume V and a dislocation density of N cm^{-2}, the fraction of atoms lying on dislocations is proportional to $NV^{2/3}$ and the mean-square displacement is proportional to $NV^{2/3}b^2$ (b is the Burgers vector). The mean-square displacement due to thermal vibrations at high temperatures is given by $\hbar^2T/(kM\theta^2)$, where θ is

the Debye temperature and M is the atomic mass. With the ideal resistivity proportional to the mean-square displacement, they obtained

$$\frac{\Delta\rho}{N} = \frac{\alpha b^2 V^{2/3} M\theta^2 \rho_i(T)}{T} \tag{2}$$

where α is a constant and ρ_i is the ideal resistance of the solid. The value of α was obtained by fitting the derived result to experimental data for copper. Based on a simple geometrical argument, they tentatively concluded that the dislocation core contributed only a minor part to the total dislocation resistivity (this conclusion did not significantly affect their results, however). The above simple semi-empirical derivation gave quite reasonable results, which had been unattainable in all of the various models developed over the previous 15 years.

THE BROWN MODEL

An adequate description of dislocation resistivity based completely on theoretical grounds was developed by Brown in 1967,[11] and subsequently modified to its final form[12,13] in 1977. By assuming a resonance in the s-wave scattering of Fermi electrons from the core of a dislocation, he was able to obtain a simple formula for the dislocation specific resistivity. Brown was able to show theoretically that resonance scattering is a characteristic property of linear defects in crystals. The basic concepts of his initial theory are outlined below.

Assuming the free electron approximation, the current density in a crystal at time t is given by

$$J(t) = -\frac{e\hbar}{4\pi^3 m} \int_{\vec{k}\ \text{space}} d^3k\ \vec{k} f(\vec{k},t) \tag{3}$$

where $(\frac{N\Omega}{4\pi^3})\ f(\vec{k},t)\ d^3k$ is the number of electrons in the crystal of volume $N\Omega$ which have wave vectors in the element d^3k of $\vec{k}$ space. Steady state requires

$$[\frac{\partial f}{\partial t}]_E + [\frac{\partial f}{\partial t}]_S = 0 \tag{4}$$

where E and S refer to the rates of change of occupation of states due to, respectively, acceleration by an electric field transverse to the defect line, and scattering by the dislocation. The first term is equal to $(\frac{e}{\hbar})\ \vec{E} \cdot \vec{\nabla}_K f(\vec{K})$ and the second term is expressed in terms of a scattering probability. The solution of Eq. (4) is substituted into Eq. (3) to give the dislocation conductivity:

$$\sigma_d = -\frac{N\Omega}{4\pi^3}\ (\frac{e\hbar}{2\pi m})^2 \int_{-\infty}^{\infty} dk_3 \int_0^{\infty} dk_* \ k_*^3\ (\frac{\partial f_o}{\partial \varepsilon})_k\ P(k_3,k_*) \tag{5}$$

where $\vec{k} = \vec{k}_* + \vec{k}_3$ and $\vec{k}_*$ and $\vec{k}_3$ are the wave vectors normal and parallel to the dislocation. The equilibrium Fermi distribution function f_o is a function only of $\varepsilon = \hbar^2k^2/2m$, and as $T \to 0$ takes the values 0 or 1 depending on whether ε is greater or less than the Fermi level ε_f. $P(k_3,k_*)$ is given by

$$P(k_3,k_*) = \{\int_0^{2\pi} W_d(k_3,k_*;\phi)(1 - \cos\phi)\ d\phi\}^{-1} \tag{5a}$$

where $(4\pi^3/N\Omega)W_d(k_3,k_*;\phi_f-\phi_i)\ d\phi_f$ is the probability of an electron in state $\vec{k}_i = \vec{k}_3 + \vec{k}_{*i}$ being scattered by the dislocation into a state $\vec{k}_f = \vec{k}_3 + \vec{k}_{*f}$. $\vec{k}_{*f}$ has its cylindrical polar angle in the range $d\phi_f$ about ϕ_f, provided that the initial state is occupied and the final state empty. A phase shift analysis of the line-defect scattering indicates that $P(k_3,k_*)$ is independent of k_3

for free electron scattering. Equation (5) then becomes

$$\sigma_d = \frac{N\Omega}{L} \frac{e^2}{4\pi^2\hbar} \int_0^{k_f} k_*^3 (k_f^2 - k_*^2)^{-1/2} \{ \sum_{m=-\infty}^{\infty} \sin^2(\eta_{m+1} - \eta_m) \}^{-1} \, dk_* \qquad (6)$$

where L is the length of the straight dislocation, k_f is the wave vector at the Fermi surface, and $\eta_m(k_*)$ is the phase shift in the mth radial function involved in the expansion of the perturbed wave function corresponding to the unperturbed plane wave of wave vector $\vec{k}_3 + \vec{k}_*$. Most of the conduction electrons in a metal have wavelengths on the order of a lattice constant (energy $\simeq$ Fermi level). Therefore, it is to be expected that the effects of the dislocation core, where the lattice distortion is varying rapidly, should dominate the electrical resistivity effects which are due to the long-range strain field.[11,14] This premise is supported by experimental evidence which indicates that dislocation electrical resistivity is insensitive to dislocation arrangement.[15] From Eq. (6) one can see that the highest resistivity is obtained for electron resonances near the Fermi energy, but that only changes in the positions or widths of the resonances which are an appreciable fraction of ε_f will affect the order of magnitude of the resistivity. For resonant scattering, the electron scattering rate is independent of the cylindrical polar angle ϕ (s-wave scattering). Therefore, a good estimate of the resistivity may be obtained from Eq. (6) by retaining only the zeroth-order phase shift. From the discussion above concerning the insensitivity of the dislocation resistivity to the positions of the resonances, one may assume $\sin^2 \eta_o$ to be a constant which is of order unity. Then Eq. (6) becomes

$$\rho_d = \beta \left(\frac{12 \pi^2 \hbar}{e^2 k_f^3}\right) \langle \sin^2 \eta_o \rangle_{av} \qquad (7)$$

where β is the density of dislocations per unit area. Values of $\langle \sin^2 \eta_0 \rangle_{av}$ may be calculated on a theoretical basis.[11] Comparison of calculated results with experimental values of dislocation electrical resistivity in several metals gave quite reasonable agreement (within a factor of two).

Brown re-examined the problem of dislocation resistivity in a series of papers[12,13] published in 1977. He deduced that low-order perturbation theory (i.e., Born approximation), which was generally used in the early theories, is not applicable for scattering by a line defect, and that a non-perturbative phase shift approach similar to his 1967 publication[11] was appropriate. He discussed in detail the following shortcomings of his original model: (1) The original theory was based on a one-band model, which is too restrictive and not applicable to transition metals. (2) The theory assumed a highly localized model of the perturbation, and it was not clear how the results depend on this approximation. (3) A simple-minded approach to the phase shift analysis was adopted -- the model considered only the zeroth-order phase shift. (4) It was not possible to make general predictions about the location of the resonance with respect to the Fermi surface, nor about the resonance width.

He addressed each of these issues as follows: (1) It can be shown[12] that the occurrence of interband scattering can only <u>decrease</u> the resonance value of the inverse relaxation time, which is proportional to the resistivity. Transition metals have a <u>higher</u> observed dislocation resistivity than theory predictions, so it may be concluded that interband scattering is not the cause of the discrepancy and may be neglected for the resonant scattering case. (2) A less-localized perturbation which gives a bias towards forward scattering, as in the case of scattering by the long-range strain field of a

dislocation, will greatly enhance the scattering cross section.[12] However, this small-angle scattering is not expected to greatly affect the momentum transfer cross-section. The localized, severely distorted core region might be expected to scatter isotropically. In this work, strain field scattering was ignored and good agreement with experiment was obtained. Therefore, it may be concluded that the contribution of strain field scattering to momentum transfer processes is negligible, and only core scattering (highly localized perturbation) need be considered. (3) For a highly localized perturbation (only one non-zero matrix element of the perturbation potential in Wannier representation), only one phase shift is present. For a moderately localized core, the symmetrized combinations of Green functions which determine the scattering are such that a resonance is only expected for the symmetrical irreducible representation.[13] It is then reasonable to assume that the s-wave phase shift dominates and is not dependent on the assumption of a highly localized potential. (4) The agreement of theory with experiment indicates that conduction electron scattering resonances from line defects occur at or near the Fermi energy, as was originaly proposed by Brown.[11] Experimental results (e.g., dislocation thermoelectric power) also tend to support this conclusion.[13]

Brown then presented an alternate derivation of the dislocation specific resistivity, applicable to multiband metals, as is outlined below.[13] Assuming a highly localized potential, the generalized expression for the conductivity tensor at T = 0 due to multiband core scattering by β parallel dislocations per unit volume is given by[16]

$$\sigma_{\alpha\gamma} = \frac{e^2}{4\pi^2\hbar^2} \sum_j \int \frac{|m_j|}{\nu_{jB} + \nu_{jD}} \{\vec{v}_{j\alpha}\vec{v}_{j\gamma} + \sum_{j'} D_{jj'} \overline{v_{j\alpha}v_{j'\gamma}}\} \, dk_3 \qquad (8)$$

where α, γ = 1, 2, 3 represent Cartesian coordinates with the dislocation lines parallel to the x_3 direction, j is the band index, m_j is the cyclotron mass for orbits along the intersection of the Fermi surface (FS) with the plane k_3 = constant, ν_{jB} and ν_{jD} are the inverse relaxation times for, respectively, background (thermal) scattering and dislocation scattering out of band j, $\vec{v}_j(\vec{k}) = \frac{1}{\hbar} \nabla_k E_{jk}$, and $D_{jj'}$ are matrix elements which weight the inverse relaxation times for dislocation and background scattering out of j into the j' band. One may verify that $\overline{v}_{j\alpha} = 0$ for $\alpha \neq 3$ as long as the contours of integration in Eq. (8) are closed (i.e., closed sheets of FS). At low temperatures in pure samples, dislocation resistivity dominates over thermal resistivity ($\nu_{jD} >> \nu_{jB}$). By assuming closed sheets of FS, the terms involving $D_{jj'}$ are equal to zero (for α, $\gamma \neq 3$) and the expression for the conductivity normal to the dislocation lines at low temperatures becomes

$$\sigma_{\alpha\gamma} = \frac{e^2}{4\pi\hbar^2} \sum_j \int \frac{|m_j|}{\nu_{jD}} \overline{v_{j\alpha} v_{j\gamma}} \, dk_3 \; ; \qquad \alpha, \gamma = 1, 2 \; . \tag{9}$$

The approximation that the contribution of open sheets of FS to the resistivity is negligible is equivalent to assuming that departures from Matthiessen's rule are not too severe.

Equation (9) can be further simplified by assuming a spherical Fermi surface. Although this certainly is not a good approximation for metals such as Mo, W, and Be, it should be rather satisfactory for the alkalis and noble metals and serves to give order-of-magnitude estimates for the other metals. It is a reasonable approximation in view of the experimental inaccuracies associated with measuring the dislocation resistivity. With this assumption, Eq. (9) becomes

$$\sigma = \frac{e^2}{\Omega_o} \sum_j \frac{n_j}{|m_j| \nu_{jD}} \tag{10}$$

where n_j is the number of carriers per primitive cell inside the jth sheet of the FS and Ω_o is the primitive cell volume. The denominator inside of the summation, for spherical sheets of FS, is given by

$$|m_j| \nu_{jD} = \frac{4\hbar\beta \sin^2 \omega_j}{d_j} \tag{11}$$

where d_j involves the ratio of the elements of the T matrix for interband scattering to those for intraband scattering $\left(\left|\frac{T_{jj'}}{T_{jj}}\right|\right)$ and is greater than or equal to one. By assuming that interband scattering is negligible compared to intraband scattering, d_j becomes equal to one.

There is empirical evidence that dislocations in single band metals have scattering resonances close to the Fermi energy. Since interband scattering is considered negligible, it is then reasonable to assume scattering <u>within each band</u> for the multiband case can be treated as if it were an independent band in a single-band metal. Therefore, it is expected that resonance scattering near the Fermi energy will occur in each band j and $\sin^2 \omega_j \cong 1$ for all j. With this simplification, the dislocation resistivity becomes

$$\rho_d = \frac{4\hbar\Omega_o\beta}{(e^2 \sum_j n_j)} . \tag{12}$$

This equation is identical to Brown's initial estimate (Eq. 7) in the single band limit. For a random distribution of dislocations, a factor of 2/3 arises from geometry considerations:

$$\left(\frac{\rho_d}{\beta}\right)_{random} = \frac{8\hbar\Omega_o}{3e^2 n} \tag{13}$$

where n is the total number of carriers per primitive cell.

A comparison of this theoretical result with experimental data has been made in Table 1. Several points are worth mentioning concerning this comparison. First, it can be seen that good agreement (generally within a factor of two) has been reached between theory and experiment for a wide range of types of metals. This builds a strong case for Brown's model since the agreement is reasonable with data that ranges over five orders of magnitude for metals from several different groups in the periodic table. There are no adjustable parameters built into the theoretical model. The agreement is particularly good for those metals such as copper which have the most reliable experimental data. Most other experimental values of the resistivity are only accurate to within a factor of two, as was noted earlier. The discrepancy between theory and experiment for the case of nickel is probably due to spin-disorder scattering, which was not accounted for in Brown's model. Second, experimental evidence indicates that the dislocation distribution is not important in determining the resistivity, which is an indication that dislocation core scattering is indeed the appropriate mechanism to consider. In general, the close agreement indicates that the approximations which were made to derive Eq. (13) are reasonable.

SUMMARY AND IMPLICATIONS

Early theories on the electrical resistivity of dislocations considered only lattice strain field scattering, and they neglected core scattering because of its small volume. The resultant calculations were generally low by more than an order of magnitude as compared to experiment. Attempts to "patch

Table 1. Comparison of Calculated and Experimental Values of Dislocation Specific Resistivity (from Ref. 13)

Metal	Structure (lattice Constant, Å)	Primitive Cell Volume Ω_0 (Å^3)	Carriers Per Primitive Cell, n	$\Delta\rho/\beta$ (10^{-19} Ωcm^3) Calculated from Equation (2.8)	$\Delta\rho/\beta$ (10^{-19} Ωcm^3) Experimental
K	BCC (5.3)	74	1.0	8.1	4
Cu	FCC (3.6)	12	1.0	1.3	1.3 ± 0.1
Ag	FCC (4.1)	17	1.0	1.9	1.9
Au	FCC (4.1)	17	1.0	1.9	2.6
Be	HCP (a = 2.3) (c = 3.6)	16	0.064	28	34
Cd	HCP (a = 3.0) (c = 5.6)	43	0.19	25	24
Al	FCC (4.0)	16	1.0	1.8	1.5 ± 0.3
Zr	HCP (a = 3.2) (c = 5.1)	45	0.13	40	100
Ti	HCP (a = 2.9) (c = 4.7)	34	0.13	29	100
Pb	FCC (4.9)	30	0.78	4.2	1.1
Bi	trig (a = 4.7) (α = 57°)	70	4.8×10^{-5}	1.7×10^{5}	2×10^{5}
Mo	BCC (3.1)	15	0.44	3.7	5.8
W	BCC (3.2)	16	0.24	7.4	7.5
Pt	FCC (3.9)	15	0.42	4.0	9
Fe	BCC (2.9)	12	0.70	1.9	10 ± 4, (2.5?)
Ni	FCC (3.5)	11	1.06	1.1	10, (2.5)
Rh	FCC (3.8)	14	1.10	1.3	36

up" the discrepancies with experimental findings included ascribing the majority of the observed dislocation resistivity to the coincident stacking faults. Refined experiments and stacking fault resistivity calculations showed that this effect was not significant. A simple semi-empirical relation based on the similarity of ideal resistivity to thermal energy provided the first reasonable agreement with experimental results. A nonperturbative phase shift approach which assumes dislocation core scattering is dominant and that there is a resonance in the s-wave electron scattering appears to model the observed results well.

This result may be applied to resistivity studies of irradiated metals. The effective resistivity of a cluster of vacancies or interstitial atoms produced during an irradiation can be reduced if the cluster collapses into a dislocation loop. Theoretical estimates of the critical size required to cause collapse of a cluster into a dislocation loop range from six[17] to 400 defects.[18] Bullough and Perrin[19] used an elastic continuum model and found that a plate-like cluster would collapse into a dislocation loop for cluster sizes greater than 22 vacancies. For a perfect loop in copper, this corresponds to a cluster diameter of ~ 11 Å. Figure 1 compares the resistivity per defect as a function of dislocation loop size for perfect and faulted loops. The effective resistivity per defect for a 50-50 concentration of vacancies and interstitials in copper is 1 μΩ-m/at. frac. defects. Therefore it is seen from Fig. 1 that there will be substantial reduction in the observed resistivity for perfect loops with diameters larger than 20 Å. It is important to account for this size-dependent loop resistivity in order to obtain accurate quantitative results from resistivity measurements, i.e. clustering can dramatically reduce the effective Frenkel pair resistivity if the cluster

collapses into a dislocation loop.

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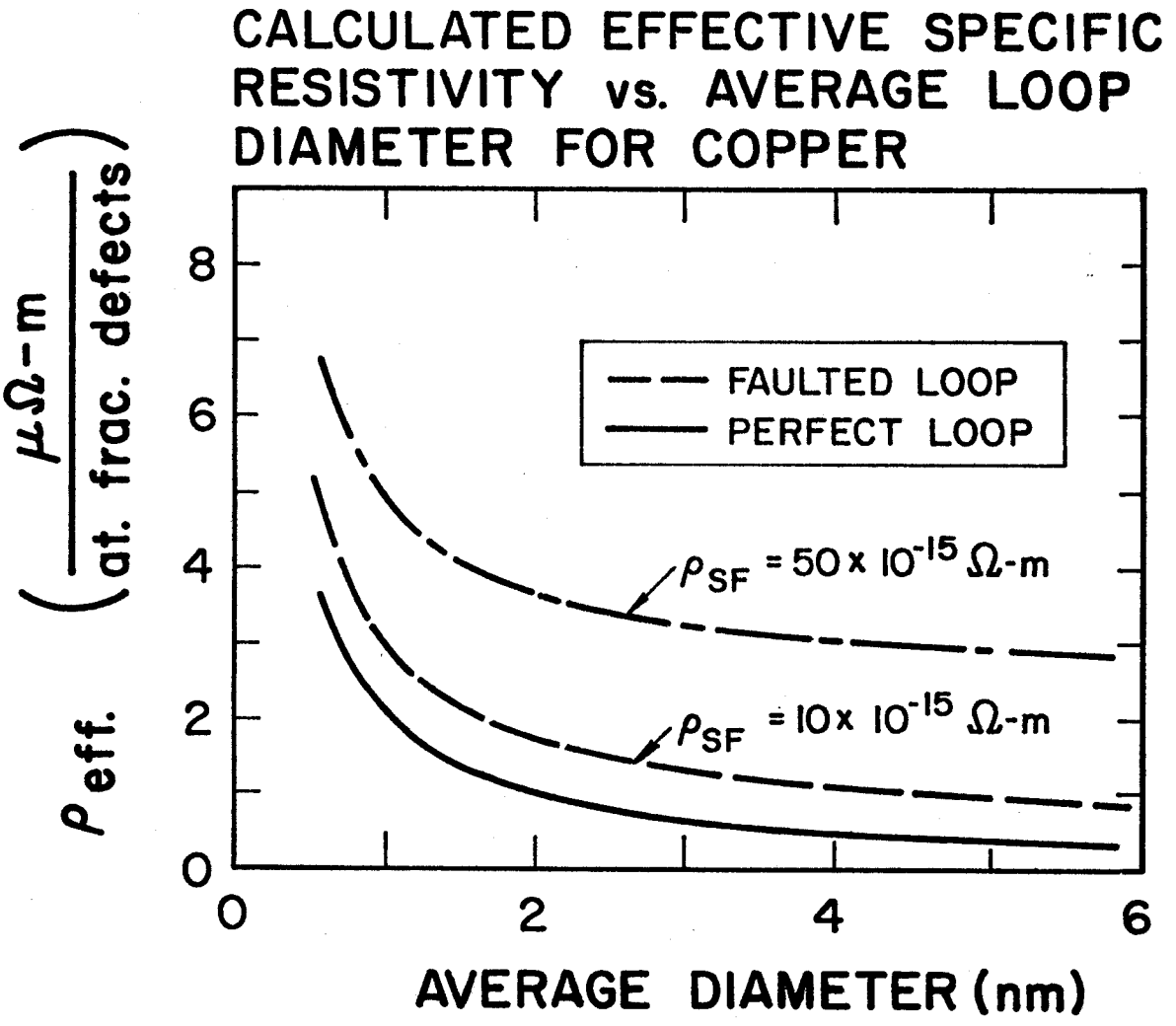

Figure 1 Calculated Effective Specific Resistivity vs. Average Loop Diameter for Copper, Assuming a Log-Normal Distribution of Dislocation Loops

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