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Conduction-electron spin resonance in cold-worked Al, Cu, and Ag: The spin-flip cross section of dislocations

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We report a study of the variation with cold working of the conduction-electron spin-relaxation time in aluminum, copper, and silver by using the transmission-electron spin resonance (TESR) technique. Measurements at low temperature of the TESR linewidth before and after proper annealing of the three metals enables us to determine the effect of dislocations on the spin-relaxation rate. From the approximately linear increase of the spin-relaxation rate with resistivity it is possible to determine a spin-flip cross section. We have compared this spin-flip cross section due to the induced mechanical defects with the well-known cross section due to thermal phonons. *For a given resistivity,* in the case of copper and silver, we found 25 ± 10% less spin relaxation for defects than for phonons. For aluminum the defects are much more effective for spin relaxation, giving four times the expected contribution. We tentatively interpret this effect on aluminum in terms of an anisotropy of the "g" factor on the Fermi surface.

I. INTRODUCTION

The study of the conduction-electron spin-relaxation time in pure metals can be considered as a study of the spin-dependent part of transport phenomena. In this sense, conduction-electron spin-resonance (CESR) measurements add to resistivity measurements to complete our knowledge of conduction-electron relaxation in metals, either spin dependent or not. Among the different possible causes of conduction-electron scattering in metals, thermal phonons and, in a great number of cases, impurities have been extensively studied in the past, both in CESR linewidth[1-5] and in resistivity.[6] Another important source of scattering in pure metals is the mechanical defects: dislocations, stacking faults, etc. A lot of work has been done on these defects concerning their effect on the electrical resistivity.[7-9] Until now, however, only qualitative studies have been made on the spin relaxation induced by such defects. The purpose of the present paper is to give quantitative data on spin scattering caused by defects introduced by cold working of aluminum, copper, and silver. We think that, besides the interest of such knowledge on a fundamental level, such a study can be useful in practice, since if one knows the effect of dislocations on the CESR signals of pure metals (or alloys), then one knows the amount of care that must be used when preparing and annealing metallic samples for CESR experiments.

In Sec. II of this paper we analyze the method we use to study the effect of dislocations on CESR linewidth in pure aluminum, copper, and silver. In Sec. III experimental procedures are described, and experimental results are given in Sec. IV. Finally, we give a brief discussion of our results in Sec. V.

II. METHOD FOR THE STUDY OF DISLOCATIONS BY CESR

In order to determine the density of dislocations existing in a cold-worked metallic sample, one can measure the low-temperature resistivity, since one knows that the increase of resistivity induced by dislocations is proportional to their density. Many attempts have been made to estimate theoretically the coefficient of this proportionality, i.e., the resistivity induced by a unit density of dislocations. One theoretical approach has consisted of only considering the long-range deformations caused by the dislocation, and neglecting the short-range core contribution. All of these calculations[10] give resistivities one or two orders of magnitude smaller than the observed ones.[7-9] This leads one to think that the major contribution to the resistivity comes from the core of the dislocation. However, only rough models of such a very disturbed region can be made, resulting in very rough estimates of the dislocation resistivity.[11]

Considering the difficulty of giving a satisfactory picture of electronic scattering by a dislocation, it seems to be very difficult to give a quantitative estimate of the spin-flip scattering induced by a dislocation. So we prefer to study the spin-relaxation rate of dislocations by comparing it with the spin-relaxation rate of thermally excited phonons. Before describing how this comparison can be made, we must recall here some CESR properties of the three metals we have studied.

Al, Cu, and Ag. There are some similarities between the properties of CESR linewidth and resistivity for metals. The resistivity can be divided into a sum of different contributions (for instance resistivity due to impurities, to crystalline defects, to sample surfaces, to phonons) assuming that Matthiessen's rule is followed. In the same manner, one can divide the observed CESR linewidth into a sum of different contributions of the same origin. CESR linewidth, like resistivity, is proportional to a probability of scattering, so one may again assume some Matthiessen's rule for linewidth, i.e., additivity of the different contributions. (We neglect at this stage of the discussion all possible effects of g-factor anisotropy on the observed linewidth; this is a good first approximation at least for copper and silver.[4,5])

The variation with temperature of the contribution to CESR linewidth due to thermal phonons has been calculated by Yafet.[12] He shows there is a proportionality between the temperature variation of CESR linewidth and that of the electrical resistivity as given by Grüneisen's law (a T^5 law for $T < \Theta$ and a T law for $T > \Theta$, where Θ is the Debye temperature). The temperature dependence of observed CESR linewidths pretty well follows Yafet's predictions.[4,5] At very low temperature, the linewidth is "residual" in the sense that it is only due to impurities, defects, or surface effects. All these effects can be, in principle, minimized, so CESR linewidth at low temperatures can be lowered to a few gauss. This statement is true for[2] Cu and[3] Ag, but Schultz *et al.*[13] observed that they were unable to obtain less than a 30-G-wide line for the purest aluminum sample at X band. More recently, a large increase of the residual linewidth of Al samples with CESR frequency was reported, first by Lubzens *et al.*[5] (9.2 and 35 GHz) and then by Janssens *et al.*[14] (21 GHz).

This frequency-dependent linewidth is not the only strange magnetic property of aluminum, compared with the more normal metals Cu and Ag, or the alkaline metals. Delafond *et al.*[15] observed a rapid variation of the total susceptibility of Al with temperature and with alloying. They report a 30% decrease of susceptibility between He and room temperature for pure Al, and some 70% decrease when dissolving 12% zinc in it.

Having recalled some CESR properties of Al, Cu, and Ag that we shall use later, we now describe how we compare dislocations and thermal-phonon effects on CESR linewidth. Taking the model of a dislocation as being built up of a long-range elastically deformed field and a highly disordered core with a diameter of the order of an interatomic distance, it is tempting to make an estimation of the contribution of the elastic deformation to the spin-flip rate by considering the long-range deformation field of the dislocation to be a sum of static phonons, and then using the experimentally known effect of thermal phonons on the spin-flip rate of conduction electrons. However, the "static" phonon wave-vector distribution one must consider to describe the long-range deformation associated with a dislocation, being proportional to $1/q$ (q is the phonon wave vector), cannot be produced by thermal excitation at any temperature.[10] So, if we try to compare CESR effects of elastic deformation and of thermal phonons at a temperature giving the same contribution to resistivity, we find a difficulty: The wave-vector distributions are quite different, and furthermore the matrix elements for scattering with and without spin flip follow a different q-vector dependence. Indeed Yafet[12] showed that the phonon wave-vector dependence for spin flip is q^2 in lowest order, whereas it varies like q for resistivity. So the effects in CESR linewidth of deformation and of phonons cannot be *a priori* compared at a given resistivity. Fortunately for our purpose, Yafet showed that spin-flip scattering and resistivity remain proportional at any temperature, owing to the different factors weighting the elementary diffusion in each case. The consequence of this is that although it is not possible to define a temperature to represent the phonon field associated with the long-range displacements around a dislocation, it is still meaningful to compare the spin-flip rate associated with it to that of a given number of thermally excited phonons of equal resistivity. Another way to see this is to realize that longitudinal phonons yield a ratio of spin-flip rate to resistivity which is independent of wave vector.[16] Once this fact is recognized, one may ask the following question: Is Yafet's result relating the spin-flip rate to the resistivity strictly valid only within the elastic limit of the displacements or can it be expected to hold for larger amplitude displacements where the elastic model breaks down, as is the case for the core of a dislocation?[17] In fact, it is this generalization which is implied more or less tacitly in the present discussion of the spin-flip rate of the dislocations, since it is well known that the main source of scattering lies in the dislocation core. However, no microscopic treatment will be given here to justify this generalization.

Now, let us see how in practice we compare dislocation and thermal-phonon effects. This comparison is made by plotting the CESR linewidth versus the corresponding resistivity induced either by dislocations or by thermal phonons. In such a plot, as we have already seen, the points corresponding to thermal phonons at different temperatures must be represented approximately by a straight line going through the origin. If we consider now our assumption that a dislocation can be analyzed in terms of a sum of static phonons, then for a given cold-worked sample, the point obtained in the ΔH-ρ diagram must fall on the phonon line. We have taken this test as a guide for the comparison of the behavior of the three metals we have studied. Even if there was no fundamental reason to expect a good fit between dislocations and phonons for a given metal in the ΔH-ρ plot, this method will enable us to see whether Al, Cu, and Ag behave in the same manner, or not.

III. EXPERIMENTAL

A. Room-temperature rolling

We have rolled at room temperature thin foils of Cu, Ag, or Al. Rolling is known to induce principally point defects and dislocations; but some of these defects will recover at room temperature, in the metals studied. Let us first consider point defects. Interstitials instantaneously recover at room temperature, because of their high mobility. Vacancies are less mobile but their density at 300 K is small; we verified this point in copper by making a partial anneal of a cold-worked sample (≈ 100 °C), which showed a very small decrease of the low-temperature resistivity, corresponding to the recovery of point defects. Thus, the majority of defects are certainly dislocations. Their mobility at room temperature depends on the metal considered.

Electron-microscopy observations of some of our samples show that in high-purity aluminum, dislocations group themselves into walls, which separate dislocations—free subgrains of diameter of the order of 1 μm in our case. However, a typical distance between two dislocations in such a subboundary is 100 Å, so that for a conduction electron (whose de Broglie wavelength is a few angstroms) dislocations are seen to be at random in the metal. This can be confirmed by resistivity measurements: At a given density of dislocations, there is a very small difference of resistivity between the polygonized and the nonpolygonized structure.[7]

In high-purity copper, on the contrary, dislocations are at random in the metal, as proved by electron microscopy. We think the same thing is true concerning silver, since in these two metals dislocations are known to dissociate into two half-dislocations separated by a ribbon of stacking fault[18]; this splitting drastically diminishes the dislocation mobility. Such a dissociation is not observed in aluminum.

B. Samples

Samples were prepared from high-purity metals: 50-μm-thick aluminum foils,[19] and polycrystalline rods of copper[20] and silver.[21] From copper and silver rods, we prepared ~ 100-μm-thick foils by rolling and annealing pieces of them. Then for each metal, the starting material was thus 50–100-μm-thick, well-annealed foils (the annealing conditions employed are given in Sec. III C). In order to introduce a more or less controlled degree of cold-working, we rolled pieces of these foils between ~ 1-mm-thick slabs of aluminum for Al samples, or of copper for Ag and Cu samples, in order to prevent pollution during rolling. We checked the degree of cold working by determining the ratio between initial and final thickness of the foil. To obtain suitable samples for transmission-electron spin resonance (TESR), i.e., samples of thickness in the 10–50-μm range, we sometimes had to use chemical thinning: dilute nitric acid for copper, and a mixture of ammonia and hydrogen peroxide for silver; no chemical thinning was needed for aluminum samples.

C. Measuring procedure

We made on each sample the following cycle of experiments: We measured CESR linewidth at low temperature (helium temperature range) and resistivity at 4.2 K. The sample was next submitted to an annealing under vacuum (using a silicon oil diffusion pump giving a residual pressure lower than 10^{-6} Torr) in order to eliminate the major part of dislocations. Typical annealing conditions were 350 °C for 15 h for aluminum or for $1\frac{1}{2}$ h for copper and silver. We found such annealing conditions sufficient to restore the original resistivity ratios of the pure metals. On each annealed sample we than systematically made CESR linewidth and resistivity measurements as before annealing. These last measurements enabled us to subtract from the corresponding values obtained on the cold-worked sample the residual contributions due to impurities, surface relaxation effects, etc. Note here that doing so assumes Matthiessen's rule for resistivity and the corresponding additivity rule for CESR linewidth.[22] The final result of such a cycle of experiments yields one point in the ΔH-ρ plot.

This complete procedure has not been followed in the case of silver, because we had substantial pollution of our samples, of unknown origin, during the annealing, which appeared as a broadening of the CESR line even though the resistivity ratio returned to its initial value. We were forced to use a unique well-annealed sample to make the above-mentioned correction on all the silver samples. We think this does not badly effect the precision of our results.

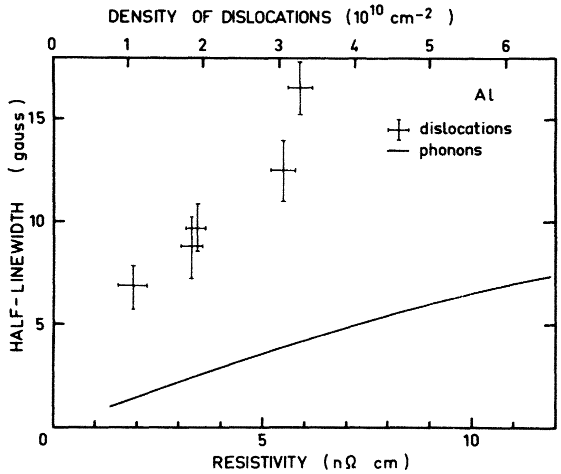

FIG. 1. Half-CESR-linewidth variation versus resistivity variation due to either phonons or dislocations for aluminum. The linewidth is taken at half-height of a symmetric Lorentzian signal. The solid line represents the contribution of thermal phonons. The points with error bars correspond to our rolled samples. An approximate scale for dislocation density, deduced from resistivity, is added.

D. Apparatus for TESR

The TESR spectrometer works in the X band (9.3 GHz). The microwave power of the order of 100 mW is amplitude modulated at 100 kHz; homodyne detection is used. A thin (typically 40 μm) foil of metal is placed tightly between an emitter and a receiver microwave resonant cavity tuned to the same frequency. The conduction electrons diffuse from one face to the other inside the foil, so that at resonance a small amount of microwave power is transmitted by the conduction-electron spins. This power (typically 10^{-16} W) radiates into the receiver cavity and is detected and amplified. Such a spectrometer has been described and discussed by Lewis and Carver[23] and by Schultz and Latham.[2]

IV. EXPERIMENTAL RESULTS

We first consider what limits the precision of our experiments. Our resistivity data were obtained by measuring resistivity ratios of samples between room temperature and liquid-helium temperature. This procedure is a very easy and rapid one and gives a typical precision of 2% on the residual resistivity. However, owing to the inhomogeneous density of dislocations over a rolled sample, a higher precision seems to be unnecessary.

Two features limit the precision of our data on CESR linewidths. In the cold-worked state, the lines are generally broad (40–200-G linewidths) so that the absolute error on full linewidths is typically 1–5 G. In the annealed state, the lines are narrower (approximately 25 G for Cu and Ag, and 50 G for Al) but a large "cyclotron" background[2] appears at low temperatures, where our measurements are performed. These signals, which depend on the resistivity of the sample, are usually one order of magnitude greater in the annealed state than in the cold-worked state. Their intensity, like the conductivity, decreases when the temperature increases so that small temperature fluctuations induce baseline drifts, reducing the accuracy of CESR linewidth determinations. The net result of this situation is that the absolute precision in the annealed state is not much higher than in the cold-worked state: typically about 1 G. Moreover, in many cases, the slight increase of resistivity caused by cold working reduces the cyclotron baseline to such an extent that the absolute precision on the linewidth can become greater, even though the linewidth increases.

The results of our experiments are presented in Figs. 1–3 on $\frac{1}{2}\Delta H$-versus-ρ plots (in TESR, when the line is symmetrical, the linewidth at half-height is equal to $2/\gamma T_2$). For each metal we have plotted the CESR linewidth versus the resistivity line due to thermal phonons, as deduced from data in the literature.[4–6] Since, for dislocations, resistivity is proportional to dislocation density, we have given on the abscissa axes two scales: resistivity (in nΩ cm) and the number of dislocations per cm^2 which we have estimated from the resistivity of our cold-worked samples, and the coefficient of resistivity per unit density of dislocations found in the literature.[7–9] Electron-microscope determinations of dislocation densities in three aluminum and two copper samples showed good agreement with these estimates from resistivity measurements.

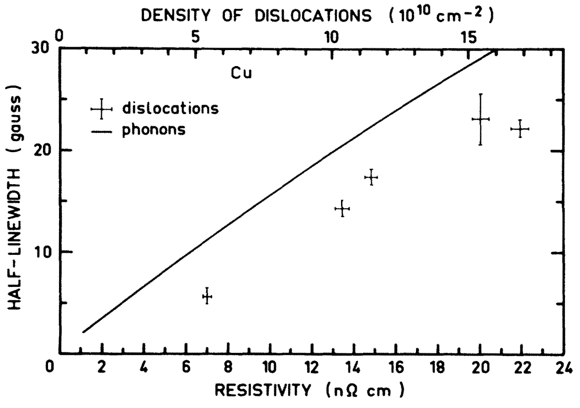

FIG. 2. Half-CESR-linewidth variation versus resistivity variation due to either phonons or dislocations for copper. A scale for dislocation density is added.

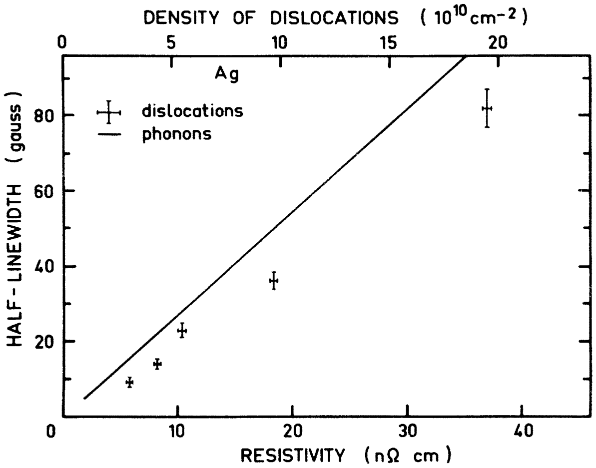

FIG. 3. Half-CESR-linewidth variation versus resistivity variation due to either phonons or dislocations for silver. A scale for dislocation density is added.

Some features are common for the three metals. (i) The points corresponding to different degrees of cold working can be practically fitted to a straight line passing by the origin of coordinates. (ii) Within experimental error, no *g* shift has been detected in the resonance of conduction electrons in the cold-worked state; *g* values always remained the same as for well-annealed metals: 1.997 for Al, 2.033 for Cu, and 1.983 for Ag (approximately ±0.003 in each case).

However, if one compares the effects of dislocations and thermal phonons, one can distinguish two types of behavior: (i) For copper and silver, dislocations pretty well follow the behavior of thermal phonons, giving only some 20% less spin relaxation than phonons for equal resistivity. (ii) For aluminum, the dislocations are much more effective for spin flip, giving approximately four times the contribution of phonons at a given resistivity.

In Table I we give an alternative presentation of these results. We list, both for phonons and for dislocations, the ratios between relaxation with and without spin flip, in two equivalent forms: first the ratios between half-CESR-linewidth and resistivity, which correspond to the slopes of the phonon or dislocation lines in Figs. 1–3; second, a dimensionless quantity corresponding to the ratio of T_2 (spin-relaxation time) and τ (resistivity lifetime), that is to say, the number of collisions a conduction electron can suffer before flipping its spin. Next we give the values of the resistivity for unit dislocation density at 4.2 K as given in the literature.[7-9] From these last values and from the $\frac{1}{2}\Delta H/\rho$ values for dislocations we can then deduce approximate values of half-CESR-linewidth for unit dislocation density, given in the last line of Table I.

V. DISCUSSION

Our experimental results show that:

(i) One can determine for the three metals a spin-flip cross section for dislocations (more rigorously a "spin-flip cross diameter," as a dislocation is a one-dimensional defect) because of the observed proportionality between CESR linewidth and resistivity which proves the proportionality between CESR linewidth and dislocation density. The coefficient of this proportionality is given in Table I.

(ii) Copper and silver have very similar behaviors. The values obtained for CESR linewidth due to dislocations are very close to the corresponding values for phonons giving the same resistivity. This close connection tends to support the conjectures made in Sec. II. One can note that for these two metals, among all known causes of conduction-electron scattering in metals, and for a given resistivity, dislocations give the least spin relaxation (with the possible exception of surface relaxation, where the data are not presently sufficient).

(iii) Aluminum results disagree with the phonon prediction, showing much more spin flip induced by dislocations than could be predicted by phonons. This disagreement between dislocations

TABLE I. Comparison between dislocations and phonons effects in CESR.

		Al	Cu	Ag	Units	Refs.
Phonons	$\frac{1}{2}\Delta H/\rho$	0.75±0.10	1.45±0.10	2.70±0.10	G/nΩcm	4–6
	T_2/τ	3900	950	350		
Dislocations	$\frac{1}{2}\Delta H/\rho$	2.6±0.5	1.12±0.15	2.2±0.2	G/nΩcm	this work
	T_2/τ	1100	1200	400		
Resistivity per unit density of dislocations		1.8±0.1	1.3±0.1	1.9±0.1	10^{-19} Ωcm^3	7–9
Half-linewidth per unit density of dislocations		4.7±1.2	1.5±0.3	4.2±0.7	10^{-10} Gcm^2	this work

and phonons, added to other strange magnetic properties of aluminum (see Sec. II) led us to think that the behavior of cold-worked aluminum in CESR might be "anomalous." In particular, the fact that pure aluminum exhibits a residual linewidth of unknown origin and that this linewidth is strongly frequency dependent, led Lubzens *et al.*[5] to suggest that there is in aluminum a broad distribution of g factors around the Fermi surface, and that this distribution is not completely narrowed by the motion of the conduction electrons, giving an important contribution to the linewidth. If one assumes such a large g-factor distribution, it is natural to think that the electrons on the Fermi surface which show a g factor quite different from 2 are precisely those electrons which show a non-free-electron-like behavior, that is to say, the electrons of the third Brillouin-zone pockets. The question is then how can a large g distribution in aluminum affect the results obtained in our measurements of dislocation-induced linewidth. In strained aluminum, the Fermi level moves relative to the energy bands, and thus the Fermi surface is deformed, as proved by de Haas–van Alphen measurements.[24] The deformation of the third zone pockets is particularly large, as a typical variation of third zone orbits areas is 1% per applied kbar. On the other hand, within a somewhat large radius around a dislocation line, very high stresses are present (a few kbar) corresponding thus to large local displacements of the Fermi level. These displacements will particularly affect the shape and dimension of the third zone pockets, to which we attribute a major part of the g-factor distribution. We suggest that it is possible that dislocations strongly modulate the g-factor distribution over the Fermi surface in aluminum, leading thus to a possible mechanism for an extra increase of CESR linewidth not due to relaxation. We do not consider the present evidence as proof of the existence of such an effect but offer this as a likely and interesting possibility.

VI. CONCLUSIONS

The case of cold-worked copper and silver seems to be simple. For a given resistivity, dislocations give a minimum contribution to CESR linewidth: This fact enables the experimentalist to use not-very-well-annealed samples for CESR measurements (getting more resistivity is often useful because the skin depth increases, and so the signal intensity increases; moreover, in TESR experiments, increasing sample resistivity diminishes to a very large extent the unpleasant "cyclotron" background, resulting in greater signal visibility). For "normal" metals, one can now, by using our method of comparing the effect of dislocations and phonons on the CESR, give an *a priori* estimate of the effect of dislocations on linewidth.

The case of aluminum is much less clear. Is the extra linewidth introduced by dislocations due to spin relaxation, or is a part of it due to a modulation by dislocations of the g-factor distribution over the Fermi surface? This suggests that it would be interesting to repeat our experiments on cold-worked aluminum at another resonance frequency.

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[16] In CESR linewidth both longitudinal and transverse pho-

nons contribute; but in resistivity only longitudinal ones contribute. So the proportionality between linewidth and resistivity will be true only if the longitudinal- and transverse-phonon spectra are similar.

[17] One experimental hint of this fact may be found in R. A. B. Devine, and R. Dupree, Philos. Mag. 21, 787 (1970): In sodium, near the melting point, the ratio between CESR linewidth and resistivity remains the same within 20% in the solid and in the liquid states. The same result qualitatively holds for potassium [see R. A. B. Devine and R. Dupree, Philos. Mag. 22, 657 (1970)].

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[19] Material Research Corp. "Marz"-grade high-purity aluminum; residual resistivity ratio ≃1000.

[20] ASARCO 99.999 +% high-purity copper; residual resistivity ratio ≃700.

[21] COMINCO 99.9999% high-purity silver; residual resistivity ratio ≃350.

[22] Surface relaxation effects are known to be quite large for resistivity [L. R. Kirkland and R. L. Chaplin, J. Appl. Phys. 42, 3054 (1971)]; for CESR linewidth no precise data are available but the effects are certainly small in our thickness range [A. Janossy and P. Monod, J. Phys. F 3, 1752 (1973)]. However, since the thickness of each sample is the same before and after annealing, ignoring surface relaxation effects is equivalent to assume Matthiessen's rule.

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