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Theory and experiment on deviations of Matthiessen's rule in dislocated crystals: a constructive reply

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Abstract. In a recent comment, Kaveh and Wiser defend their theory on deviations of Matthiessen's rule (DMRs) by questioning related experiments of ours on aluminium, because of inconsistencies in experimental data as well as fatal influences of an extended size effect. The present reply points out essential errors with both criticisms and thus strongly insists on the validity of the experiments originally reported. The DMRs are far smaller than predicted by Kaveh and Wiser. Nevertheless, owing to some correlation of data in literature with deformation mode and annealing treatment, the DMRs appear to arise from long-range strain fields close to the dislocations. This suggests that the Kaveh–Wiser theory could apply for particularly strain-intensive dislocation arrays and help to quantify the dilatation-specific part of dislocation resistivity.

1. Introduction

Since the pioneering work of Hunter and Nabarro [1], the electrical resistivity ρ^d of dislocations has been studied continuously. In that paper, and in the major part of the work done up to now, ρ^d has been thought to arise mainly from the elastic dilatation field of the dislocations [2–6], which causes a *strongly anisotropic small-angle scattering* of electrons (these models are hereafter referred to as models A).

However, exact measurements of the *specific* dislocation resistivity ρ^d/N (N is the dislocation density) exhibited a large discrepancy between experimental data and models A; the experimental value of ρ^d/N exceeded the theoretical value by two orders of magnitude.

In recent years, the problem has been re-treated on contrary assumptions (hereafter referred to as models B). Here, an idea of Harrison [7] has been gradually developed, in which the scattering of a dislocation was simulated by a linear row of vacancies. As a consequence, these models imply a rather *isotropic scattering* behaviour of mainly *large-angle* type. An alternative method of description was given by Brown [8], who discussed possible effects of such missing atoms on the resulting band structure (similar to effects of dislocation in semiconductors), and the possibility of additional localised states close to the Fermi surface in the vicinity of dislocations [8]. Under the assumption of an s-wave resonance scattering, calculation of ρ^d/N for 17 metals was achieved and were in accordance with experiments to within a factor of 2 [9].

In spite of the conciseness of this introduction, we should discuss the most important consequences of the two models.

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(i) *Models A* clearly imply that ρ^d/N depends not only on the density N but also on the arrangement of dislocations. In contrast with this, *models B* have ρ^d/N rather insensitive to different dislocation arrangements as long as N is the same, i.e. low-strain structures with stress-intensive pile-ups of dislocations and large-strain structures of cells and/or subgrains which are poor in local internal stresses should yield identical values of ρ^d/N; in the extreme, the experimentally obtained values of grain-boundary resistivity should be correctly calculable on the basis of well known models of narrowly spaced dislocation patterns forming the grain boundary [10].

(ii) Superposition of cooperative electron scattering from dislocations with that from other lattice defects (i.e. phonons and impurities) should affect the actual ρ^d/N-value in cases where the anisotropy of respective scatterers differs significantly. Such contributions are typically not constant in temperature and well known as deviations from Matthiessen's rule (DMRs) in the literature. For the above reason, they should emerge even more drastically in models A than in models B, where the role of the more anisotropic scatterer is thought to be played by the dislocations involved.

Concerning point (i), only a few experiments exist (parallel observations by electrical resistivity and by TEM, [11–13]). Several investigations, however, were made with respect to (ii), DMRs in connection with dislocations as a function of measuring temperature, although there emerged marked differences in the nature and extent of DMRs observed [14–19]. Furthermore, there has been reported a series of theories [20–22] which predict such distinct DMRs in case of coexisting isotropic *and* anisotropic scatterers, the most extensive having been formulated by Kaveh and Wiser [22, 23]. Here, satisfactory agreement with related experiments could be gained for the case of cooperative impurity and phonon scattering [22]. Large discrepancies, however, emerged in connection with additional dislocation scattering [18, 23], especially if one claims a mainly anisotropic small-angle scattering of electrons by dislocations.

The present paper defends previous experimental data of ours obtained from Al as a function of dislocation density and phonon contribution [13, 18] and proves all objections which were raised in a recent comment by Kaveh and Wiser [24] to be wrong. Moreover, further experimental data from previous literature are presented which confirm our findings of a rather *isotropic* scattering behaviour of dislocations, in contradiction to the results of Kaveh and Wiser once more. However, a few experiments which achieved extraordinarily high partitions of dilatation field either caused by the particular deformation mode or by the small amount of strain applied seem to be described satisfactorily by the above theory. Thus, this paper finally discusses the conditions under which the strain-field contribution would become significant to ρ^d/N and suggests possibilities for quantifying it; then, it should be possible to give *separate* values for the specific core resistivity $(\rho^d/N)_{core}$ and for the specific dilatation-field resistivity $(\rho^d/N)_{strain}$, which would help to avoid confusion and irreproducibility with both theoretical and experimental considerations.

2. Defence of experimental data

2.1. Relationships between dislocation resistivity ρ^d, dislocation density N and strain ε

A few years ago, we performed careful measurements of ρ^d in pure Al [18]; the results showed that the theory of Kaveh and Wiser [23] does not describe (ρ^d/N) (N) correctly, especially in the range of dislocation densities $N \leqslant 10^{10}\,\mathrm{cm}^{-2}$. In a

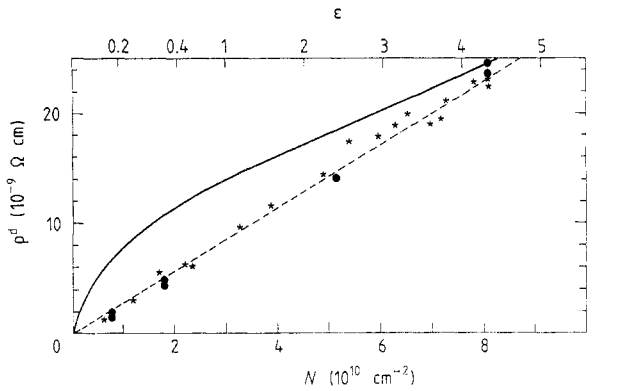

Figure 1. The total dislocation resistivity ρ^d in Al at 77 K, as a function of dislocation density N, and true strain $\varepsilon = \ln(d_0/d)$ (d is the sample thickness [13]). The deviation of values measured by our group (*, [13]; ●, [18]) from the Kaveh–Wiser theory [18, 23] (——) is apparent. Note the drastic change in ε scaling at $\varepsilon \simeq 0.4$.

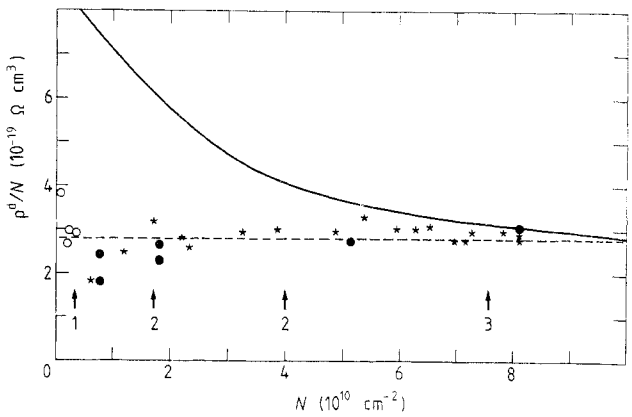

Figure 2. The specific dislocation resistivity ρ^d/N in Al at 77 K, as a function of dislocation density N: *, [13]; ●, [18]; ○, [11]. The arrows indicate how the respective N data have been obtained: arrow 1, direct measurements by TEM [11]; arrow 2, data from literature on TEM (see also [13]); arrow 3, calculation from TEM measurements [13], and/or from constancy of $(\rho^d_{4.2K}/N)(N)$ in this range of N, according to [23].

recent comment, Kaveh and Wiser [24] called into question these experiments, since they would not be consistent with previous experiments of our group [13], which, moreover, would agree well with their theory. However, this assertion is not true; it essentially arises from the incorrect assumption of Kaveh and Wiser that the relation between dislocation density N and the strain ε is *linear* (see, e.g., figure 1 of their work [24]) which, however, is only true for strains $\varepsilon > 0.4$ in the form $N = c'\varepsilon + c''$, and certainly wrong for the low N-values (found as a roughly parabolic relation $N \sim \varepsilon^2$ [25]). This range exhibits a markedly larger slope [13] and is far more important for the present discussion. In figure 1, the results from our experiments in [13] and [18] have been plotted in the correct way. The reproduction of these values is quite satisfactory which means that the deviation of experiments from the theory is *unique*. Analogous to the comment of Kaveh and Wiser, we also give a plot ρ^d/N against N (figure 2), consisting of the theoretical curve of Kaveh and Wiser, of (correctly plotted) experimental results of our group [13, 18] and of those of most reliable other work [11]; here, strong differences from the theory are evident, once more justifying the conclusions drawn in our paper [18]. The experimental results quoted by Kaveh and Wiser which fit their calculation well (see figure 2 of their comment [24]) are of doubtful value. Apart from the disadvantage that they do not span the whole interesting range of relevant N-values, they both stem from investigations of quenched-in loop dislocations which easily lead to certain overestimations of ρ^d/N. First, numerous

'loops' could be too small for identification in the electron microscope [26] and/or actually could consist of vacancy clusters; secondly, the nature of the loops has not always been identified uniquely [14, 26, 27], possibly yielding certain contributions of stacking-fault resistivity. Anyway, owing to inexact experimental procedures, Yoshida *et al* revoked their older findings [28] which have been so strongly referred to by Kaveh and Wiser [24]. They reported an upper limit value of $(\rho^d/N) = 1.5 \times 10^{-19}\,\Omega\,cm^3$ in the following paper [26], which lies close to our measurements [13, 18].

2.2. Influences from the temperature dependence of size effect

Kaveh and Wiser [24] also strongly doubt our previous measurements [18] owing to a marked temperature-dependent size effect contribution ρ^s to dislocation resistivity ρ^d which they quote as arising both from the limited thickness of samples and from variations therein. They refer to the recent measurements given in [29] which fit well to a law $\rho^s \sim T^2$. However, its validity is only applicable up to temperatures of about 20–30 K; it has been clearly shown by the measurements reported in [30], which were also used in [29], that ρ^s drastically decreases beyond 30 K. Obviously, this fact has been ignored by Kaveh and Wiser when they extrapolated the above law to 77 K, arriving at rather excessive values for ρ^s of 6 nΩ cm. When estimating the possible errors from size effect correctly, the work (figure 4) of [30] yields $\rho^s = 0.7$ nΩ cm for a sample thickness d of 75 μm. For the smallest strains applied, this would still lie in the order of brought-in dislocation resistivity; however, in [30] very-coarse-grained ultra-pure material ($\rho_{4.2K} = 0.1$ nΩ cm) was used, giving an electron mean free path about 50 times larger than with our samples ($\rho_{4.2K} = 5.4$ nΩ cm). Because in [30], the found sample thicknesses d were 1 mm or more to make ρ^s less than 0.1 nΩ cm, the same value of ρ^s should apply to $d \simeq 20$ μm at the purity of samples used by us [18]. Considering their thicknesses reported above, these lie *far beyond* this value, making any size effect correction strongly negligible.

2.3. Consistency with other experimental results

The results of the most reliable investigations can also be incorporated in this proper plot of p against the residual dislocation resistivity (figure 3(*a*)). This concerns the measurements in [14] after rolling, straining and even annealing to definite values of dislocation density N, and the measurements in [10] in which TEM images were evaluated to obtain the correct values of N. They all lie markedly below the respective Kaveh–Wiser graphs, although they even used distinctly thicker samples than we did. It is interesting to note that even in Cu, where Kaveh and Wiser predict increases in p to p-values of 4–6 (figure 3(*b*)), in [12] no similarly large increase was found in spite of careful N measurements by TEM. However, the experiments in [12] do not really cover the whole range of N in question, nor do other investigations [5, 15], so that at present no final judgment of the experimental situation in Cu can be given.

3. Discussion and comments

3.1. The temperature dependence of ρ^d/N in the light of existing theories

The Kaveh–Wiser theory is also capable of yielding the whole temperature dependence of ρ^d/N. T is introduced via the temperature dependence of β, which

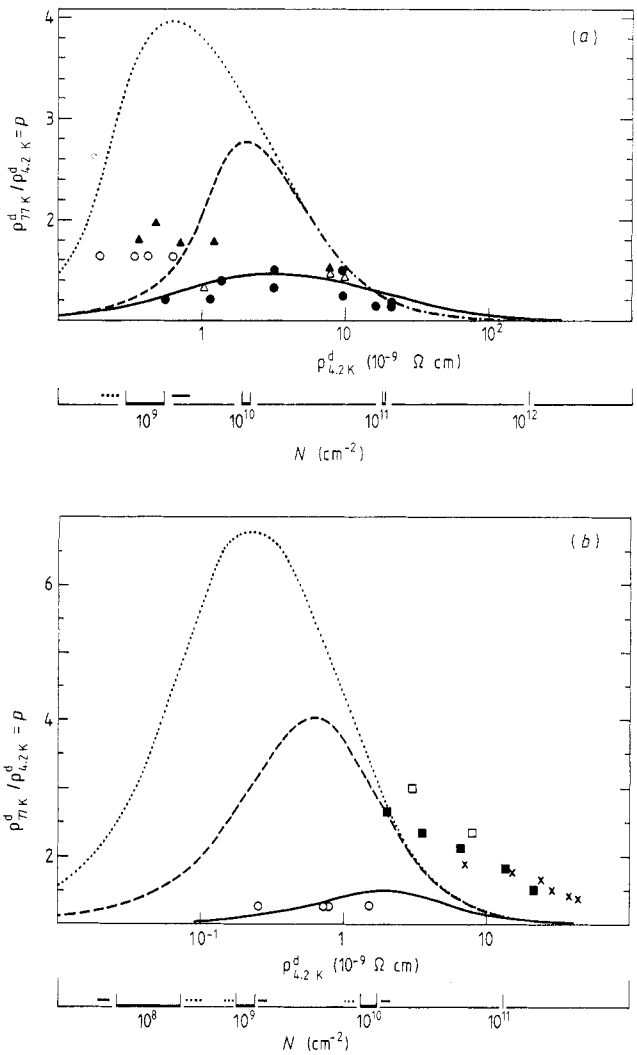

Figure 3. Ratio p of the dislocation resistivity (ρ^d_{77K}) measured at 77 K to the dislocation resistivity $\rho^d_{4.2K}$ measured at 4.2 K (as a measure of DMR) plotted against the dislocation resistivity $\rho^d_{4.2K}$ (as a measure of average dislocation density N). The curves have been calculated from the theory of Kaveh and Wiser [23] for two purities: ····, 99.999% pure; – – –, 99.99% pure. The maximum of p decreases with decreasing parameter α (decreasing anisotropy of dislocation scattering) and increasing impurity (for details see [18]). The symbols indicate representative experimental data (experiments have been performed on polycrystals, if no other type of crystal mentioned). (*a*) Al (●, 99.99% pure, rolling [18]; ○, 99.99% pure, tensile test [11]; ▲, 99.999% pure, tensile test and rolling ($\rho^d_{4.2K} > 1$ nΩ cm) [14]; △, 99.999% pure, rolling and annealing [14]; ——, best fit of data in [18]; (●) by the Kaveh–Wiser theory with $\alpha = 0.6$, $\beta = 0.1$ (77 K) and $\beta = 0.4$ (4.2 K)). (*b*) Cu (○, 99.99% pure, tensile test [12]; □, 99.999% pure, single crystals, rolling, bending [15]; ■, 99.999% pure, rolling, bending [15]; ×, 99.999% pure, wire drawing [5]; ——, best fit of data in [12] (○) by the Kaveh–Wiser theory with $\alpha = 1.0$, $\beta = 0.075$ (77 K) and $\beta = 0.3$ (4.2 K)).

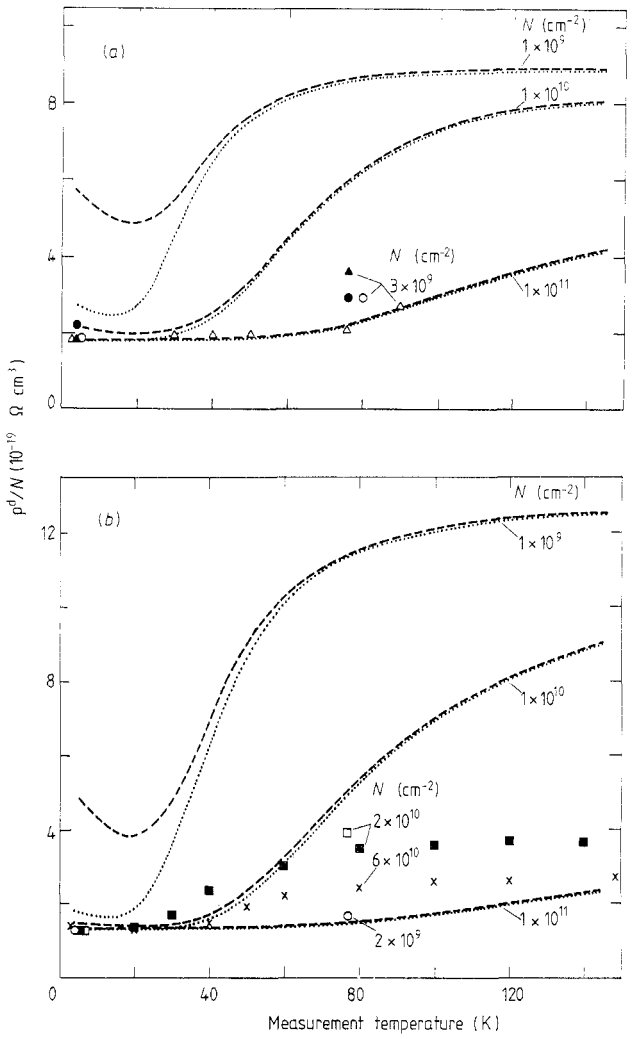

Figure 4. Specific dislocation resistivity ρ^d/N at various measuring temperatures T. For the meanings of the symbols, see figure 3. The curves result from calculations according to the theory of Kaveh and Wiser [23, 31] for purities of 99.999% (····) and 99.99% (– – – –), by assuming marked anisotropy of dislocation scattering: (*a*) Al, $\alpha = 4$; (*b*) Cu, $\alpha = 9$. If α is taken as 0.6 (Al) or 1.0 (Cu), the lowest curve results for each material, regardless of purity and/or dislocation density.

describes the non-additive scattering behaviour of the dislocations while changing the fraction of phonons. To realise the graphs of ρ^d/N against T as given in figure 4, β has been gained from respective calculations by Bergmann and co-workers [31], and its temperature dependence was taken from earlier calculations given by Kaveh and Wiser [22] for DMRs in general. As a result, while the theory exhibits strong DMRs, i.e. a marked dependence of ρ^d/N on T, the experiments do *not*. This is most spectacular in [14] with specifically annealed samples which had a residual dislocation density of about $3 \times 10^9\,\mathrm{cm}^{-2}$ (figures 3(*a*) and 4(*a*)). Moreover, in the low-temperature range $4.2\,\mathrm{K} \leqslant T \leqslant 20\,\mathrm{K}$, calculations according to Kaveh and Wiser show a slight minimum in ρ^d/N against T (figure 4), which is obviously *not* reflected in experiments on *specifically annealed* samples [14, 19, 32]. However, slightly deformed samples especially after wire drawing showed some tendency to DMRs, including several observations of the minimum mentioned [16, 19]. An interpretation of these effects will be given in § 3.2.

3.2. A constructive comment on the nature of scattering process by dislocations

Focusing on two representative experiments in Al [14, 19] by the same Japanese group, quite interesting features are exhibited as follows. Very slightly strained samples indeed exhibit such DMRs which are at least qualitatively similar to those predicted by Kaveh and Wiser; they manifest as

(1) a *negative* DMR with a minimum of ρ^d at about 10 K [19] and
(2) a *positive* DMR with a maximum of ρ^d at about 60 K and above [14].

Although both types of DMR have never been measured with the same set of samples and selective annealing treatment, nor has there been undertaken any parallel investigation by TEM giving direct structural evidence, the above results suggest that the DMRs arise from some specific contribution of the *strain fields* of introduced dislocations to the value of ρ^d/N in question.

(*a*) Heavily deformed samples in principle show markedly less total dilatation per unit dislocation density N since the drastically growing number of dislocations is arranged into low-energy dipoles (in cell walls) with mutual screening of long-range strain fields (see, e.g., [33]).
(*b*) Annealed samples imply recovered structures consisting of small- or even large-angle boundaries which do not exhibit any strain field at all [33].

This picture is also confirmed by measurements on Cu [5, 15] and Au [17] which showed a relatively large DMR of type (2), and also by experiments on Al [16, 19], Ag [16, 32] and Cu [34] which gave a DMR of type (1), for the following reasons. All these experiments have in common that *particular modes of deformation* have been applied such as *bending* [15], *wire drawing* [5, 16, 17, 19, 34] and *swaging* [32] which are predestined to form strain-field-rich dislocation arrays (pile-ups), compared with, for example, stretching and particularly rolling. Additional evidence for this explanation is given by the de Haas–van Alphen measurements in [6] performed on *bent* samples which misled Kaveh and Wiser to their identification of dislocations as *small-angle scatterers*, which obviously is *not generally* valid. It therefore seems very sensible to assume that this *small-angle scattering* arises from the dislocation's strain field (*small* atomic displacement), whereas *large-angle scattering* occurs at the dislocation core (*large* atomic displacement).

Interestingly, it seems improbable that the DMRs arise from some anisotropic scattering as a consequence of the linear dimension of the dislocation core solely. This has been shown experimentally in [35] by measurements of ρ^d vertically (to give ρ_v^d) and parallel (to give ρ_p^d) to the highly preferred direction of strain-field-free dislocation dipoles in fatigued specimens. Thereby a ρ_v^d/ρ_p^d-value as small as 1.1 or less resulted. Furthermore, it has been shown very recently [36] that a similar value can also be gained by a thorough theoretical treatment.

In the literature, sometimes other theories have been applied to explain dislocation-caused DMRs.

(i) A proportionality of $\rho^d \sim -\log T$ below the temperature of the minimum in ρ^d was found in [19] and it was thus suggested to explain the DMR of type (1) by the spin-dependent Kondo effect and/or mechanisms associated with the two-level system (TLS).

(ii) In [15, 17], attempts were made to explain the maximum of ρ^d at about 60 K, i.e. the DMR of type (2). Brown's theory [8, 9] of a dislocation-caused additional energy resonance level for the electrons was extended so that this level

should be occupied thermally by a Boltzmann probability; the respective fits to experiments gave a reasonable level difference of about $\Delta E = 10\,\text{meV}$ to the Fermi level.

The above suggestions can be briefly judged in the sense that *none* of them satisfies both types of DMR simultaneously. So, the spin-Kondo/TLS model fails in describing the 'hump' of ρ^d at 60 K, while Brown's model does not reflect at all the low-temperature minimum of ρ^d. Most important, none of the models is capable of explaining the disappearance of both type (1) and type (2) DMRs for highly deformed and specifically annealed samples.

4. Conclusions

As a consequence of the above considerations, we repeat our suggestion from [18] that the parameter α should be reduced, which somewhat reflects the actual scattering angle of dislocations [23], from $\alpha = 4.0$ to about $\alpha = 0.6$ in Al, for deformation structures with a *low* fraction of strain field (e.g. in highly deformed samples and recovered or partially annealed samples). We additionally suggest a value of about $\alpha = 1.3$ for deformation structures with an *extensive* fraction of strain field (e.g. in slightly tensile strained samples, α following from fits [37] of related data given in [11] and [14]). Values of $\alpha = 4.0$ as originally proposed by Kaveh and Wiser [23] only hold for deformation structures with *very large* strain fields, e.g. those achieved by slight wire drawing. This we conclude from numerical fits [37] of the type (1) DMR measured in [16, 19] which also account for its disappearance after selective annealing with N constant ($\alpha = 0.6$), although the contribution of impurities used in this calculation had to be taken as $6\,\text{n}\Omega\,\text{cm}$ instead of a nominal $1\,\text{n}\Omega\,\text{cm}$. In fact, it would be necessary to ensure the *kind of specific dislocation structure*, before establishing certain values for α and consequently ρ^d/N. In our opinion, the description could be markedly facilitated if one defined separate values for the dislocation core resistivity ρ^d_{core} and the strain-field resistivity ρ^d_{strain}, where both seem to be accessible by theory as well as by experiment. With respect to experiment, we suggest selective annealing treatments of the samples investigated intermittently by resistivity measurements and parallel TEM imaging, the latter in order to monitor the number and arrangement of dislocations. To quantify the contribution of the strain field, calculations based on TEM images and/or parallel calorimetry measurements of the deformation-induced stored energy should be carried out.

Acknowledgments

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