N O R T H W E S T E R N U N I V E R S I T Y

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Dear Editor and Referees,

Thank you for taking the time to review our manuscript on the interface scattering model for twist boundaries and semicoherent heterointerfaces.

In response to the reviewer comments, we have added statements to clarify each of the following topics in the appropriate sections of the paper:

*Section I. Introduction:*

* Dislocation strain mainly scatters mid-frequency phonons with wavelengths on the order of the dislocation spacing. High frequency phonons will interact with the dislocation core.
* Our handling of the dislocation strain fields is applicable for grain boundary angles of less than 15°C, where there is not significant dislocation core overlap.

*Section II. Theoretical Formalism:*

* The temperature dependence of the model comes explicitly from the spectral heat capacity because other parameters are largely temperature independent.
* The implementation of our model is not polarization specific, and so complex mode conversions at the boundary are not evaluated here. Phonons in this model are described by a Born-von Karman dispersion with three degenerate branches parameterized by an average speed of sound.
* To avoid issues with a changing reference lattice, phonon directions and scattering perturbations are defined with respect to a virtual average lattice.
* Details about the phonon transitions (Section IIA) and scattering potentials (Section IIB) follow after the more general model framework. We added more referencing to these sections to make the organization of the manuscript clearer.

*Supplementary Section S6:*

* The anticipated temperature dependence of the input parameters (summarized in Table S2) has a negligible impact on the predicted relaxation times. This helps to confirm that the temperature dependence of the thermal resistance is mainly controlled by the spectral heat capacity.
* We added a summary of the computational demands in terms of run times and hardware used.

We have also reordered portions of the Introduction such that the scattering potential is introduced before the discussion of the momentum transfer at the interface.

We have added a citation to the work of D. Wolf [*Scripta Metall.* **23,** 1713- 1718 (1989)], which discusses the dislocation core overlap in high angle grain boundaries, where the linear elasticity methods applied in our model begin to break down. We have also added a citation to a work by Carruthers [Carruthers, *Phys Rev* **114**, 4 (1959)] with a detailed discussion of the anharmonic, elastic scattering from a static dislocation strain field. Finally, we added a citation to [Carrete *et al., Comp Phys Comm* **220**, 351-362(2017)] as an example of method, which defines an interface scattering perturbation from a virtual average crystal.

Our responses to each question, comment, and suggestion are detailed below. Here the reviewer comments are shown in purple with our responses following in black.

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Report of the First Referee -- BA13861/Gurunathan

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In this work, the authors introduce a novel formalism for studying

phononic interfacial thermal resistance at low angle grain boundaries

and heterointerfaces. Previous models rely on differences in the bulk

properties of the phases surrounding the interface. However, this new

approach expands these models by providing additional information

concerning the local environment near the interface by incorporating

the strain field of the dislocation into the model. After describing

the model, the authors then apply it to study Si grain boundaries and

Si-Ge heterointerfaces.

Molecular dynamics studies tend to be more general due to the absence

of any assumptions on the nature of phonon scattering at the

interface. However, the fact that this model enables the user to more

explicitly control what effects are included in their analysis makes

it more informative. Generally, I found the paper to be very

well-written and I feel the work is an important advancement in the

understanding of interfacial thermal phenomena. Therefore, I would

like to recommend this manuscript for publication in Physical Review B

provided the following minor comments and concerns are addressed.

1) The authors demonstrated that the model works well in the low-angle

grain boundary regime where the dislocation cores are relatively

sparsely spaced. However, what is the range of applicability of the

model? Presumably, for large enough misorientation angles the model

will stop working? Can the authors provide an estimation for the

maximum misorientation angle for which this model can be expected to

yield accurate results?

Our treatment of the interfacial strain fields involves 1) using the Read-Shockley model to relate the grain boundary angle to the interfacial dislocation spacing and 2) computing the interfacial strain field by performing a periodic sum over the single dislocation strain fields described using continuum elasticity theory. As noted by the reviewer, both of these strategies are applicable when there is not significant dislocation core overlap, as in low-angle grain boundaries [Cai and Nix, *Cambridge Uni. Press,* pp. 688-700(2016)]. Dislocation core overlap typically begins around 15° in most materials [Wolf, *Scripta Metall.* **23,** 1713- 1718 (1989)]. We confirm this to be true in our Si grain boundary examples by noting that the interfacial energy begins to plateau at 15°.

We have added a statement to the paper noting that the model is expected to yield accurate predictions for grain boundary angles less than 15° in most materials. Additionally, we have added a citation to the work of D. Wolf describing the implications of dislocation core overlap at high grain boundary angles [Wolf, *Scripta Metall.* **23,** 1713- 1718 (1989)].

2) The model requires several input material parameters, as referenced

in Table S2. These parameters are generally functions of

temperature. Did the authors do any analysis to estimate the

sensitivity of the model predictions with respect to typical

variations in these parameters? If the variation is non-negligible,

then in the future one could express these parameters as functions of

temperature to generalize the model. For example, by using data

obtained from quasi-harmonic density functional theory calculations.

Thank you to the reviewer for suggesting this interesting extension to the current model. As suggested, we have checked the sensitivity of the model to small changes in these input parameters. To do so, we have used the lattice parameter change due to thermal expansion of silicon from 300 to 800°C (approximately 0.18% increase in lattice parameter) and propagated this effect to the density and speed of sound. The resulting change in the computed phonon relaxation times was only about 0.16%, so it seems like temperature dependence of these input parameters is a negligible effect. We have added a statement to Supplementary Section S6 noting this, but also suggesting, as the reviewer mentioned, that temperature dependent inputs could be determined from a technique like quasi-harmonic density functional theory.

3) The temperature T appears in the expression for the spectral heat

capacity (S10). As far as I can tell, this is the only place that it

explicitly appears in the model? Unless perhaps it arises elsewhere

implicitly? In either case, it would be informative to add a sentence

specifying how the temperature was "controlled" since it appears as a

variable in several plots.

As observed by the reviewer, temperature explicitly enters into this model solely through the spectral heat capacity. We added the following sentence to confirm this in the paper: “The transmissivity and phonon velocities are often temperature independent, and so the temperature dependence of enters solely through the spectral heat capacity.” Additionally, we cite the review by Monachon *et al.* [*Ann. Rev. Mater. Research* **46,** 433-436 (2016)], which supports this point about the temperature dependence of .

4) If possible, it would be nice to provide the reader with some sense

of the computational demands required to run the model (perhaps in the

supplementary material). Specifically, what are typical run-times for

generating results, and what type of hardware was used (i.e. laptop vs

supercomputer). Such information would be useful for readers

interested in using the model to study a wider range of materials, for

instance, in a high-throughput manner.

Thank you for this suggestion. The most computationally intensive part of the algorithm is computing the integrals over both the phonon incident angle and the phonon frequency. We have expanded Supplementary Section S6 to cover the computational demands of the model as well.

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Report of the Second Referee -- BA13861/Gurunathan

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This paper develops a theoretical approach to compute phonon

scattering at interfaces. Both dislocation strain fields and harmonic

mismatch are considered.

I have some questions about the approach which I think require a major

revision. I will list the specific questions below:

1. On page 4 the point is made that strain fields induce

anharmonicity. I think that might be misleading. It definitely would

be true that the strain fields would lead to shifts in phonon

frequencies and this is related to the Gruneisen parameter. But the

actual scattering considered in the paper appears to be entirely

harmonic (i.e. energy conserving). This is a minor point but it is

confusing.

As noted by the reviewer, the scattering processes we consider are energy-conserving and are therefore elastic rather than inelastic, which we note in the Introduction section. However, the origin of the strain scattering is still anharmonic, since strain would not scatter phonons in a purely harmonic solid. We use the anharmonic Grüneisen parameter to calculate the lattice energy perturbation due to strain.

The important distinction between elasticity/inelasticity and harmonicity/anharmonicity can be understood by considering the 3rd order, anharmonic term in the Taylor expanded interatomic potential. Similar to the treatment of 3-phonon processes, the anharmonic potential for strain scattering is based on three displacement vectors (*u*). For the case of strain field scattering, one of these displacement vectors represents the static strain field which is induced by the defect [Carruthers, *Phys Rev* **114**, 4 (1959)]. The scattering is still elastic because we treat a static strain field, which lacks the dynamical degrees of freedom to absorb or emit energy (discussed in the Introduction section of the manuscript). We additionally cite Carruthers 1959 and a reference by Klemens [Klemens, *Proc. Phys. Soc. A.* **68**, 1113 (1955)], both of which discusses how static strain fields lead to elastic, anharmonic phonon scattering. In the Introduction, we also contrast our analysis to recent work on localized interface mode coupling, in which inelastic scattering is possible by transferring energy to localized vibrations at the interface.

2. Why is Eq. 1 using H^{\prime}, but then Eq. 3 is using V(r). This

seems like a minor point, but I think the notation should be consistent.

Although both represent lattice energy perturbations, we use distinct notation for and to highlight that one is a reciprocal space entity and the other is a real space entity. Conceptually, is a 3D real space function that relates directly to the deformation at the interface, while is a Hilbert space operator that relates more closely to specific phonon transitions from to . After considering the reviewer’s suggestion of using a single notation, we feel that denoting them separately will lead to less overall confusion. Equation 3 highlights their relationship through a Fourier transform.

3. I don't understand the point about choosing V(r) depending on

whether the momentum transfer Q parallel to the surface is nonzero or

not. It would seem to me that the perturbing potential is first

chosen, and then the scattering rate and momentum transfers are

determined, not the other way around.

The reviewer is correct that the differences in momentum transfer emerge from the specified scattering potential of the interface. The existence of momentum transfer parallel to the interface plane () is simply a method to distinguish the two parts of the scattering potential (the dislocation strain grid and the acoustic mismatch) based on the phonon transitions they can produce. Only the dislocation strain grid varies in the interface plane and can induce a nonzero . In contrast, only the acoustic mismatch can induce a scattering process with . In our implementation of the model, this criterion allows us to discern which component of the scattering potential to apply (dislocation strain or acoustic mismatch).

This manual modification of the scattering potential is required to keep the mathematical scattering theory consistent with the real defect structure. Figure 2, which shows the mathematical description of the interfacial dislocation strain field, highlights the necessity of this step. Figure 2 shows finite strain persisting infinitely far away from the boundary, which is an unphysical boundary condition. As a result, the far-field deformation at the interface (in Figure 2, represented by the lattice parameter change) should be treated separately from the localized, periodic strain at the boundary, and is modeled here as a source of acoustic mismatch. Our treatment differs from other recent publications [Varnavides *PRB* **100**, 115402 (2019)]. We believe this subtle, yet important point represents a unique advancement of this work.

To make this point more clear in the manuscript, we have reordered this portion of the discussion in the Introduction section, such that the scattering potential is introduced before momentum transitions are discussed.

4. Is Eq.2 only describing back-scattering? I have a question about

how the momentum transfers are used. Specifically, do the wave vectors

used in Eq. 1 relate to phonon modes on the same side of the

interface? Or opposite sides of the interface? It is confusing. I

understand that the momentum transfer will be related to the

periodicity of the dislocation array, but I'm unsure about how the

matrix elements relate to phonon modes on the left/right sides of the

interface.

As summarized by the reviewer previously, the perturbation potential contains two major parts: 1) the acoustic mismatch and 2) the dislocation strain. For the acoustic mismatch potential, Equation 2 is limited to mirror-like back-scattering, since the only alternative is forward scattering, which does not contribute to thermal resistance. Therefore, in Eq. 2, the phonon modes and are both defined on the same side of the interface (the incident side).  However, for the dislocation strain potential, there is no such limitation to just back-scattering. In this case, the matrix element may relate phonon modes on the left and right side of the interface. To avoid issues with a change in reference frame, the wavevector directions are defined in reference to a virtual average lattice, which symmetrizes the step function scattering potential from acoustic mismatch. This type of virtual crystal approach is used when treating point defect perturbations and superlattices [Carrete *et al., Comp Phys Comm* **220**, 351-362(2017)]. We have added a statement to Section IIB and a reference to Carrete 2017 to highlight this point.

5. Phonon modes are indeed characterized by wave vectors, but there

are also LA and TA modes (I believe here the focus is on relatively

long-wavelength modes, since short wavelength modes might begin to

probe the dislocation cores. The paper does not make a mention of

that. I would think that matrix elements for any perturbation would

have to account for the particular polarization of the phonon modes.

For example, are transmissions LA--> LA? Or TA--> TA? Or can LA-->TA

occur, or even the possibility of the strain fields yielding two

phonons with different frequencies (but still conserving energy)? The

latter can occur in temperature-induced strain fields, so why not in

strain fields due to strain fields due to dislocation arrays?

Thank you to the reviewer for noting this clarification that should be emphasized in our work. We have added a statement to make it more explicit that this interfacial strain scattering model is relevant for mid- to long-wavelength phonons with wavelengths on the order of the dislocation spacing or larger.

Additionally, this model is not polarization specific, and therefore does not treat the mode conversions mentioned by the reviewer. We have added a short paragraph to Section II on the Theoretical Formalism to emphasize this point. The phonons in our model are described by a Born von Karman model with three degenerate branches parameterized using an average speed of sound (values listed in Table S2). In this aspect, the implementation of our model is similar to implementations of established analytic theories such as the acoustic mismatch model and the diffuse mismatch model [Monachon, *Ann. Rev. Mater. Research* **46,** 433-436 (2016)]. However, we offer an important advance by treating interfacial dislocation strain within our framework.

Finally, similar to previous works on interfacial strain scattering [Varnavides *PRB* **100**, 115402 (2019) and Meng *PRB* **87**, 064102 (2013)], our analysis is limited to cubic anharmonic terms, which relate two phonon displacement vectors to the strain field displacement. Higher order interactions mentioned by the reviewer, which may involve three different phonon modes interacting via the strain field, have thus far been neglected. Additionally, as mentioned previously, the static strain fields lack the dynamical degrees of freedom to absorb or emit energy such that the scattering we consider is fully elastic.

6. Usually the application of the Fermi golden rule applies to weak

perturbations. However, in this case, the entire crystal can be

rotated. Can one treat a disruption in the lattice symmetry as a weak

Perturbation?

We acknowledge that the limit of what constitutes a “weak perturbation” is an open debate in the following statement in the Introduction section:

“*We acknowledge that the limits of perturbation-type methods and their application to phonon-grain boundary or phonon-interface interactions is an open debate. We hope this work demonstrates the utility of this approach.*”

We feel that the predictive power of our model suggests that this extension of Fermi’s golden rule is suitable, as suggested by another recent study [Varnavides *PRB* **100**, 115402 (2019)].

In Supplementary Section S2, we justify our use of perturbation theory even for the long-range deformation at these interfaces. We compare the phonon transmissivity results from our perturbation theory treatment to that of the Landauer theory-based acoustic mismatch model (AMM). We find that perturbation theory agrees with the AMM within 5%, even when the relative change in phonon velocity across the interface is as much as 50% (see Figure S1). Since, the phonon velocity mismatch present at most solid-solid interfaces is well below this limit, perturbation theory is expected to be suitable.

I think some description and justification along the lines above

should are considered before I could recommend publication. I think

the description of what modes are considered, or how they are defined,

starting with Eq. 1, needs to especially be considered. I do not quite

see how the perturbing potential nor the phonon modes are defined such

that the matrix elements considered in Eq. 3 make sense. There is a

lack of a clear definition of what is being done and precisely how it

makes sense.

Thank you to the reviewer for the recommended clarifications. We have added statements to the Introduction and Section II on the Theoretical Framework to better clarify our implementation of this model. Additionally, we would like to note that in the organization of the Theoretical Framework section, the high-level procedure is described first, followed by further details describing the phonon transitions and the perturbation potentials. We do this to provide a general template before delving into the specific cases for the dislocation strain and acoustic mismatch scattering potentials. However, these scattering potential and phonon mode definitions are required to fully implement Equation 3. We have added more referencing to these later sections to make this clear.