Crystalline coherence length effects on the thermal conductivity of MgO thin films

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Phonon scattering in crystalline systems can be strongly dictated by a wide array of defects, many of which can be difficult to observe via standard microscopy techniques. We experimentally demonstrate that the phonon thermal conductivity of MgO thin films is proportional to the crystal coherence length, a property of a solid that quantifies the length scale associated with crystalline imperfections. Sputter deposited films were prepared on (100) silicon and then annealed to vary the crystalline coherence, as characterized using x-ray diffraction line broadening. We find that the measured thermal conductivity of the MgO films varies proportionally with crystal coherence length, which is ultimately limited by the grain size. The microstructural length scales associated with crystalline defects, such as small angle tilt boundaries, dictate this crystalline coherence length and our results demonstrate the role that this length scale dictates the phonon thermal conductivity of thin films. Our results suggest that this crystalline coherence length scale provides a measure of the limiting phonon mean free path in crystalline solids, a quantity that is often difficult to measure and observe with more traditional imagining techniques.

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Defect-induced deviations in lattice structure can give rise to phonon scattering processes and changes in the phonon thermal conductivity of crystals. Where ample experimental works have studied and validated classical scattering theories regarding phonon-grain boundary and phonon-impurity thermal resistances on bulk- and nano-scales,1-8 the phonon scattering mechanisms contributing to thermal resistances at finer scale defects, such as small angle tilt boundaries and dislocations, have been much less frequently studied.9 This has led to voids in the depth of understanding of the interplay between, and importance of, phonon-lattice defect scattering relative to the interaction of phonons with other static impurities, such as incoherent grain boundaries, mass impurities, and interfaces between dissimilar materials. Progress in this fundamental understanding of the phonon-lattice defect interaction will have major impacts in the design of novel classes of material systems, such as the recently discovered systems synthesized by utilizing screw dislocations,10-14 high figure of merit thermoelectric materials designed with dense dislocations arrays,15 nanostructures with dislocation dense interfaces that impact the thermal boundary conductance,16-18 and thermal transport in ferroelectric materials where coherent ferroelastic domain walls affect the phononic resistance.19-21

Here, we report on measurements of the room temperature thermal conductivity of a series of magnesium oxide (MgO) nanocrystalline thin films in which the crystalline coherence lengths (the characteristic length of crystal devoid of translational symmetry-breaking defects) of the MgO films are varied. Small angle tilt boundaries defining crystallites of similar dimensions to the measured crystal coherence length were previously identified in identically processed films. These, in addition to grain boundaries and other crystallographic defects, such as dislocations, were attributed to a damping of optical phonons.22 It is therefore anticipated that these same
defects may affect the transport of heat-carrying phonons. We use time domain thermoreflectance (TDTR)\textsuperscript{23} to measure the thermal properties of the MgO thin films at room temperature and atmospheric conditions; by utilizing a combination of both the in-phase and the ratio of the in-phase to out-of-phase components of the TDTR response in tandem, we demonstrate the ability to measure the thermal conductivity of the MgO films at a single modulation frequency, while separating the influence of thermal boundary conductance across both the front and back thin film interfaces from this thin film thermal conductivity measurement. We show that the thermal conductivities of the MgO films increase with an increase in crystalline coherence length, which is correlated with not only the defects that limit the crystalline coherence, but also is indicative of a changing average lattice parameter of the MgO. Our work demonstrates the ability to quantify the influence of defects on the phonon thermal conductivity by an average length scale of crystal translational symmetry – the crystalline coherence length. Our results suggest that this crystalline coherence length scale provides a measure of the limiting phonon mean free path in crystalline solids, a quantity that can be determined via standard X-ray reflectivity and is often difficult to measure and observe with traditional microscopy techniques.

The 80 nm thick MgO film series was prepared on (100)-oriented silicon substrates via 30° off-axis RF magnetron sputter deposition within a Kurt J. Lesker Lab 18 instrument. Prior to loading in the load-locked sputter chamber, the substrates underwent a 7:1 buffered HF etch (pH of 5.5) and de-ionized-H\textsubscript{2}O rinse to remove the native silicon dioxide surface. The film was sputtered from a single-phase sintered MgO target in 5 mTorr of argon at room temperature with a power density of 3.7 W/cm\textsuperscript{2}. The wafer was subsequently divided and sections were processed between
200°C and 800°C in 200°C intervals in air for one hour; varying the annealing temperature directly correlates to a change in crystalline coherence length. Silicon was chosen as a substrate due to its predicted phase stability and chemical inertness with MgO in this temperature range, suggesting the formation of clean interfaces with no secondary phases during the deposition and annealing process; furthermore, the high thermal conductivity of silicon ensures maximum sensitivity to the thermal conductivity of the MgO thin films in our TDTR measurements. Phase purity and crystalline coherence lengths were characterized via X-ray diffraction (XRD) using a Philips X’Pert MPD with Cu Kα radiation in the Bragg-Brentano geometry. As such, the diffraction vector is normal to the sample surface and the only lattice planes to which we are collecting diffracted X-rays are those that are parallel to the film surface (actually, parallel to the 001 planes of silicon, to which the instrument was aligned, but for all intents and purposes is parallel to the surface). Therefore, crystallite dimensions to which this technique is sensitive are those normal to the film surface – the same direction as thermal conductivity is measured. Crystalline coherence lengths were calculated using X-ray line broadening and Scherrer’s formula.

The surface roughnesses, thicknesses, and density of the annealed samples were characterized using a combination of atomic force microscopy (AFM), X-ray reflectivity (XRR) and variable-angle spectroscopy ellipsometry (VASE). Cross-sectional and plan-view microstructural images (Fig. 1a–f) were obtained by scanning electron microscopy with an FEI Verios. Secondary electron images were collected with an electron landing energy of 500 V and 2000 V stage bias. Average grain sizes were determined from the plan-view images using the linear intercept method. Additionally, 78 nm thick aluminum films were electron-beam evaporated onto the
samples for use as transducers in subsequent TDTR scans. The precise aluminum film thicknesses were determined using both mechanical profilometry and picosecond acoustic measurements during subsequent TDTR scans.\textsuperscript{27,28}

As shown in the SEM images in Figure 1 a-f, polycrystalline grains with columnar morphology comprise the films. Figure 1g shows the coherent scattering lengths of X-rays for the MgO samples processed between room temperature and 800°C, and reveals a trend of increasing length with post-deposition processing temperature, similar to that reported previously in identically processed MgO thin films.\textsuperscript{22} Scattering lengths were calculated using the 200 MgO X-ray reflection; the measured coherence lengths varied from 5.4 nm for the as-deposited sample to 19.5 nm for the 800°C processed sample. Lattice parameters were also determined from 200 peak position using the silicon 400 reflection as a reference for sample displacement error correction. Shown in Figure 1h, the lattice parameter is reduced with increasing coherence length. The data indicate that the higher the degree of crystalline perfection, the more closely the lattice parameter approaches the accepted single crystal value of 4.21 Å.\textsuperscript{29} In these fine crystallite size films, it is likely that disorder near surfaces, defects, and grain boundaries gives rise to these expanded lattice parameters, as has been suggested previously for fine-grained MgO.\textsuperscript{30} SEM images of the sample series shown in Fig. 1a–f indicate that physical grain size, as defined by the distance between well-defined grain boundaries, is constant among the sample series, where the average grain size is 20.9 ± 1.3 nm, indicating that the varying crystalline coherence length is not driven by grains separated by large angle boundaries. Additionally, no obvious change in density or porosity was observed with annealing condition. No statistically significant variation in film thickness was identified (average of measurements from XRR and VASE, and listed in Table I),
further suggesting that film density was constant among the sample series. However, increasing the annealing temperature did lead to changes in the MgO surface roughnesses and the formation of increasingly thick SiO$_2$ layers between the MgO and silicon substrate with increased temperature. More specifically, the MgO surface roughness increased from 2.8±1.2 nm for as-sputtered samples to 9.3±5.3 nm for the sample annealed at 800°C (average of measurements from XRR, VASE and AFM, and listed in Table I); also, we detected an increasingly thick SiO$_2$ layer between the MgO and silicon that grew to as thick as 6.1±1.2 nm after the 800°C anneal (average of measurements from XRR and VASE, and listed in Table I). We note that the relatively large uncertainties in our reported average values for SiO$_2$ thicknesses could be indicative of the different sensitivities of XRR and VASE to this buried SiO$_2$ film, as the uncertainties are determined from the standard deviation among the data collected with each technique. Regardless, the temperature dependent variations in our samples were confined around the top and bottom MgO film interfaces. Therefore, since the microstructural properties in the MgO films away from the film boundaries are comparable among all of the films, this suggests that the only factor varying within the bulk of the film is the crystalline coherence length, which has previously been verified.$^{22}$

The thermal conductivities of the MgO films were determined using TDTR by fitting the data to a multi-layer thermal model described in detail in the literature.$^{31-34}$ Briefly, TDTR is a non-contact optical pump-probe technique that uses a short-pulsed laser to both produce and monitor modulated heating events on the surface of a sample. The laser output from a sub-picosecond oscillator is separated into pump and probe paths, in which the relative optical path lengths are adjusted with a mechanical delay stage. The pump path is modulated to create a frequency
dependent temperature variation on the surface of the sample, and the in-phase and out-of-phase
signals of the probe beam locked into the modulation frequency of the pump were monitored
with a lock-in amplifier. Prior to TDTR measurements, the sample surfaces were coated with a
thin aluminum film so that the changes in reflectivity of the surfaces were indications of the
change in temperature within the optical penetration depth of the aluminum; this change in
reflectivity is driven by the thermal properties of the MgO film, the silicon substrate, and the
thermal boundary conductances across the Al/MgO and MgO/Si interfaces. We assume
literature values for the heat capacities of the aluminum,\(^{35}\) MgO,\(^{36}\) and silicon,\(^{37}\) leaving the
unknowns in our thermal models as the thermal boundary conductances across the Al/MgO and
MgO/Si interfaces (\(h_{K,Al/MgO}\) and \(h_{K,MgO/Si}\), respectively) and the thermal conductivity of the MgO
film, \(\kappa_{MgO}\).\(^{31}\)

Typical TDTR analyses on thick films or substrates can analyze the ratio of the in-phase to out-
of-phase signals from the lock-in amplifier to determine the thermal conductivity and thermal
boundary conductance across the metal/sample interface (assuming a relatively high thermal
effusivity). For example, Fig. 2a shows a typical TDTR data set collected on our samples, with
the inset showing the corresponding thermal model fit. However, in the case of our MgO thin
films, our TDTR measurements are also sensitive to the thermal boundary conductance across
the MgO/Si interface, even at relatively high modulation frequencies (~10 MHz). Therefore, we
cannot use the ratio signal alone to measure the thermal conductivity of the MgO since we
cannot uniquely separate this from the two thermal boundary conductances. To overcome this
experimental limitation, we modify our analysis approach by utilizing a combination of both the
in-phase signal and the ratio of the in-phase to out-of-phase components of the TDTR response
in tandem. The in-phase component of the lock-in frequency response is sensitive to the Al/MgO interface during the first nanosecond of a TDTR scan, since the in-phase component is related to the single pulse response in the time domain, as shown in Fig. 2b. Exploiting this sensitivity, we fit the data for the front-side boundary conductance using the real component, then apply this value when using the ratio to fit for both the back-side conductance and MgO thermal conductivity. As shown in Fig. 2c, the ratio is highly sensitive to the thermal conductivity of the MgO and the thermal boundary conductance at the MgO/Si interface. While the ratio is only minorly sensitive to the thermal boundary conductance across the Al/MgO interface, this sensitivity can vary based on the thermal conductivity of the MgO, which changes by a factor of ~3 among the films studied in this work. Therefore, this approach is necessary to accurately measure the thermal conductivity of the MgO, while also evaluating the corresponding uncertainty in our measurements. We discuss this approach in more detail, including sensitivity analyses in our previous work.38

We use this aforementioned analysis approach in an interactive fashion, we measure $h_{K,Al/MgO}$, $h_{K,MgO/Si}$, and $\kappa_{MgO}$ at a single TDTR scan at a single frequency. To ensure accuracy of this approach, we measure $h_{K,Al/MgO}$, $h_{K,MgO/Si}$, and $\kappa_{MgO}$ at different pump modulation frequencies. Our measured results of the thermal conductivities and thermal boundary conductances are shown in Figs. 3a and b, respectively, for two representative samples (those with the largest and smallest coherence lengths: 800°C annealed and as-deposited samples, respectively). We measured $h_{K,MgO/Si}$ to be relatively constant across all the samples (~200 – 300 MW m$^{-2}$ K$^{-1}$), indicating the negligible influence of the change in SiO$_2$ thickness at the MgO/Si boundary on $h_{K,MgO/Si}$. We find no statistically significant and appreciable change in the thermal conductivity
or boundary conductances with varying frequency, indicating the robust ability of our approach to measure the intrinsic thermal conductivity of thin films with a single TDTR measurement at one modulation frequency when thermal boundary conductance could influence the thermal response. This elucidates a unique analysis procedure when using TDTR to measure the thermal properties of thin films.

Our reported uncertainties in the values reported for thermal conductivities and thermal boundary conductances are determined by considering three different sources of error. First, we calculate the standard deviation among the entire set of measurements for each sample (multiple measurements on each sample). Second, we assume a ~10% uncertainty in the Al transducer film thickness. Finally, we determine a 95% confidence interval for each measurement. We take the square root of the sum of the squares of each deviation from the mean values resulting from these sources of uncertainties to construct our error bars. We note that largest uncertainties in our reported values lie in the samples with the highest thermal boundary conductances and highest thermal conductivities. This is consistent with the fact that as the thermal conductivity of the MgO thin films increase (or the interfaces conductances increase), and hence, the corresponding thermal resistances decrease, our TDTR measurements become less sensitive to these thermophysical properties. However, our reported values still lie within a 95% confidence bound. Along these lines, it is worth noting that the apparent observed frequency dependence in the thermal conductivity measurements of the MgO samples with the maximum coherence length (Fig. 3a) are nearly constant when considering our aforementioned confidence interval, and still only deviate ~20% about the mean; in other words, this fluctuation in our measured data for thermal conductivity with frequency is not physical, but just an artifact of the sensitivity of
TDTR for measuring relatively thermally conductive thin films (i.e., films with relatively low thermal resistance), especially when using lower pump modulation frequencies where the thermal penetration depth is increased and therefore sampling more of the underlying substrate relative to the thin film. However, as the coherence length in the MgO thin film is decreased, and the thermal conductivity is lowered, TDTR measurements are much more robust and sensitive in measuring thermal conductivity, consistent with the relatively minor uncertainty associated with our fits.

Measuring the thermal conductivity as a function of crystallinity, we find that thermal conductivity varies with the coherence length and plateaus as the crystalline coherence length approaches the average grain size (20.9 ± 1.3 nm), as depicted in Fig. 4. The thermal conductivity begins to plateau at larger crystalline coherence lengths because the coherence length of these samples are ultimately limited by the large angle grain boundaries. This is consistent with Matthiessen’s Rule, which asserts that the shortest phonon scattering length scale will dominate the average mean free path. Therefore, the large reduction in the measured thermal conductivities of these MgO samples compared to bulk single crystalline MgO (Ref. 39) are due to the grain boundaries for the largest coherence lengths and limited by various imperfections in the crystal as the coherence length is decreased. Because the film thickness, density, grain size, and porosity are consistent among the sample series, the only factor that changes in these polycrystalline films is the crystalline coherence length, further supporting this observation of a transition from imperfection-limited thermal conductivity at small coherence lengths to grain boundary limited thermal conductivity at the larger coherence lengths.
We note that the surfaces of the MgO film are changing among the samples processed at different temperatures; as previously mentioned, with increasing MgO processing temperature, the MgO surface becomes more rough and the SiO\textsubscript{2} layer between the MgO and silicon becomes thicker. However, we measure an increasing thermal conductivity with increased temperature, which would imply that the increased surface roughness and increase in SiO\textsubscript{2} thickness, which would add thermal resistance to the system, play only a minor role in our thermal conductivity measurements of MgO compared to the changing crystalline coherence length. This also gives further support to our data representing the intrinsic thermal conductivity of the MgO, and our ability to separate the resistances at the MgO interfaces from our reported values of $\kappa$.

To put the magnitude of the reduction in thermal conductivity due to the crystalline coherence length scales into perspective, we turn to the minimum limit to thermal conductivity.\textsuperscript{40} Assuming an isotropic solid, the minimum limit is given by

$$\kappa_{\text{min}} = \frac{\hbar^2}{6\pi^2 k_B T^2} \sum_j \int_0^{\omega_{c,j}} \frac{\omega^4}{\tau_{\text{min},j}} \frac{\exp\left[\frac{\hbar \omega}{k_B T}\right]}{\exp\left[\frac{\hbar \omega}{k_B T}\right] - 1} \frac{d\omega}{v_j}$$

(1)

where $\kappa_{\text{min}}$ is the minimum thermal conductivity, $j$ is the phonon polarization index, $\tau_{\text{min}}$ is the minimum scattering time, $\omega$ is the angular frequency, $\omega_{c,j}$ is the cut-off frequency, and $v_j$ is the phonon group velocity. To evaluate Eq. (1) for this material system, we use a Debye assumption with sound velocities of the acoustic branches taken from the experimentally determined dispersion\textsuperscript{41} in the [100] direction. While the lowest thermal conductivity sample is a factor of 4 higher than the thermal conductivity predicted from the minimum limit, it is lower than the bulk thermal conductivity by an order of magnitude.\textsuperscript{39}
To confirm that this variation in thermal conductivity is due to the change in crystalline coherence length, and not simply a result of changing lattice parameter, we compare our experimental results to the Leibfried-Schloemann equation\(^{42}\) given by
\[
\kappa = \frac{\beta M a \theta_D^3}{T \gamma^2}
\] (2)
where \(\beta\) is a factor inversely related to \(\gamma\) (the Gruneisen parameter), \(M\) is the average atomic mass, \(a\) is the lattice constant, \(\theta_D\) is the Debye temperature, \(T\) is temperature, and \(\gamma\) is the Gruneisen parameter, given by
\[
\gamma^2 = \frac{a^6}{\omega^2 \left( \frac{\partial \omega}{\partial (a^3)} \right)^2}
\] (3)
We determine \(\partial \omega/\partial (a^3)\) from the experimentally measured transverse optical frequencies, which depend on unit cell volume\(^{22}\) and scale the Debye temperature for each film assuming \(\theta_{D,\text{film}} = \theta_{D,\text{lit}}(a_{\text{film}}/a_{\text{lit}})\). Through this analysis, we observe completely different trends when comparing our experimental results to the predicted variation of thermal conductivity due to lattice spacing. This implies that the change in lattice parameter among the sample series is not responsible for the variation in thermal conductivity, rather, that defects responsible for the crystalline coherence are the driving force impacting phonon scattering.

This demonstrates the ability to quantify the influence of defects on the phonon thermal conductivity the crystalline coherence length of the crystal. This has the advantage of offering limiting length scales for phonon transport in crystalline system in which imperfections are difficult to characterize and/or model. For example, using molecular dynamics simulations, Ni et al.\(^9\) showed that localized strain field, varying atomic spacing, and modifications to the intrinsic anharmonic phonon-phonon interaction strength near defects, such as dislocation cores, must be accounted for to properly model the phonon-lattice defect dynamics in the thermal conductivity.
Unlike phonon-grain boundary and phonon-mass impurity-limited thermal transport,\textsuperscript{1-8} modeling these processes is not easily or accurately feasible with simplified kinetic theory-type models, and therefore predictions of changes in thermal conductivity due to these imperfections can be daunting. However, our work suggests that the characterization of a crystalline coherence length gives insight to qualitatively compare changes in thermal conductivity of similar materials with different degrees of crystalline imperfections.

As a final note, our experimental measurements in Fig. 3b show a relatively negligible dependence of thermal boundary conductance with crystalline coherence length of the MgO. We have previously observed that interfacial imperfections can lead to changes in thermal boundary conductance.\textsuperscript{16,43} Given that we do not observe any substantial structural changes at the surfaces of the MgO films, we would not expect any changes in thermal boundary conductance, which is consistent with our measurements of $h_K$ at each interface. Furthermore, it interesting to note that the Al/MgO thermal boundary conductance is consistently lower than the MgO/Si thermal boundary conductance regardless of the MgO crystalline coherence length. While more work must be done that specifically focuses on the role of interface defects, our results highlight the potential impact of our previously discussed TDTR analysis to extract the thermal boundary conductance across thin films interfaces, and using this to assess the role of changes in atomic-scale defects at material interfaces on changes (or lack thereof) in thermal boundary conductance.

In conclusion, we have investigated the effects of crystallinity changes on thermal conductivity of MgO thin films. We find a systematic increase in thermal conductivity with increasing
coherence length. Our thermal model, while sufficient for many other material systems and phonon scattering processes, fails to account for this crystallinity effect. This is consistent with previous studies, and implies that much more complex modeling is necessary to understand the effects of dislocations on phonon scattering.

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References


Table I. MgO film thickness (average of measurements made with XRR and VASE), MgO surface roughness (average of measurements made with XRR, VASE and AFM), and thickness of SiO₂ layer between MgO and Si substrate (average of measurements made with XRR and VASE). The uncertainty reported in these measurements represent the standard deviation among all the values determined from the different techniques.

<table>
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<th>Anneal temperature (°C)</th>
<th>N/A</th>
<th>200</th>
<th>400</th>
<th>600</th>
<th>800</th>
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<tr>
<td>MgO film thickness (nm)</td>
<td>84.0±2.8</td>
<td>78.9±4.9</td>
<td>79.3±3.5</td>
<td>83.6±1.7</td>
<td>82.4±1.6</td>
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<tr>
<td>MgO surface roughness (nm)</td>
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<td>5.8±3.4</td>
<td>4.72±1.8</td>
<td>5.1±2.3</td>
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<td>3.4±3.1</td>
<td>3.5±2.1</td>
<td>6.1±1.2</td>
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Figure 1: SEM images for varying processing temperatures: (a) 25°C, (b) 200°C, (c) 400°C, (d) 600°C, (e) 800°C; (f) cross-section for representative sample; (g) coherence length as a function of temperature; (h) lattice parameter as a function of coherence length. The SEM plan view images show that grain size and porosity are consistent among all of the films, and the cross-sectional images show that all films are 80 nm thick. The coherence length is found to vary in direct proportion to the processing temperature. As the crystallinity increases, the measured lattice parameter decreases and approaches the single crystal value intrinsic to MgO.
Figure 2: (a) Typical TDTR data on our MgO films for the largest coherence length sample (blue line/higher data set) and lowest coherence length sample (black line/lower data set); (inset) corresponding thermal model fits to the largest coherence length sample’s representative TDTR
Sensitivities of our thermal model for determining the thermal boundary conductances across the Al/MgO and MgO/Si interfaces ($h_{K,Al/MgO}$ and $h_{K,MgO/Si}$, respectively) and the thermal conductivity of the MgO ($\kappa_{Si}$) from analyzing the (b) in-phase and (c) ratio of in-phase to out-of-phase TDTR data.
Figure 3: (a) Thermal conductivities and (b) thermal boundary conductances as a function of modulation frequency. Analyzing the in-phase signal in tandem with the ratio of the in-phase to out-of-phase signal, we find that, within a standard deviation, the thermal conductivities and thermal boundary conductances for the maximum and minimum coherence lengths are constant among varying modulation frequencies. This is confirmation that our analysis technique enables us to determine $h_{K,\text{Al/MgO}}$, $h_{K,\text{MgO/Si}}$, and $\kappa_{\text{MgO}}$ from a single TDTR scan at any given modulation frequency.
Figure 4: Thermal conductivity as a function of coherence length. The bulk value for MgO given from Touloukian et al. is shown for comparison.\textsuperscript{39} We observe an increase in thermal conductivity with increasing crystalline coherence length, which is ultimately limited by the grain size of the polycrystalline MgO films. The reduction in crystalline coherence length leads to MgO samples with thermal conductivities that are roughly a factor of 4 higher than the predicted minimum limit (Eq. 1), a factor of 3 lower than the largest coherence length sample (limited by grain boundary scattering, which remains constant for all coherence lengths), and a factor of 10 lower than bulk MgO.\textsuperscript{39}