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Transport properties in β-Ga_2O_3 thin films

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Novel Materials Group, HU Berlin

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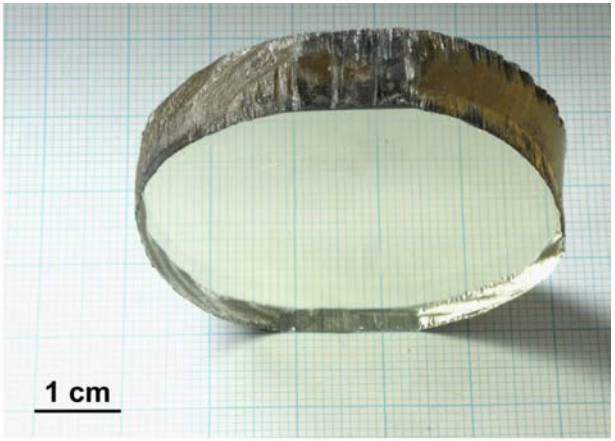
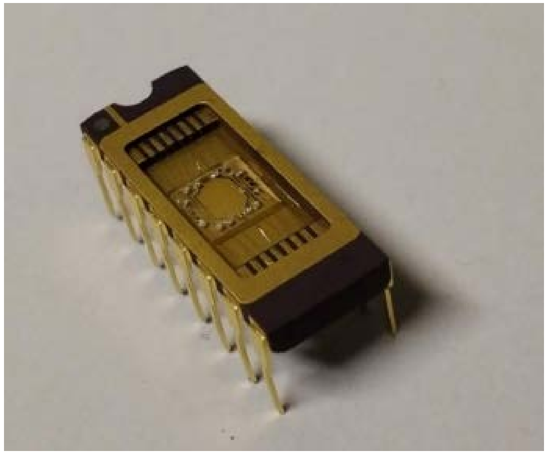




DFG
FI 932/10
FI 932/11

Leibniz Institute for Crystal Growth

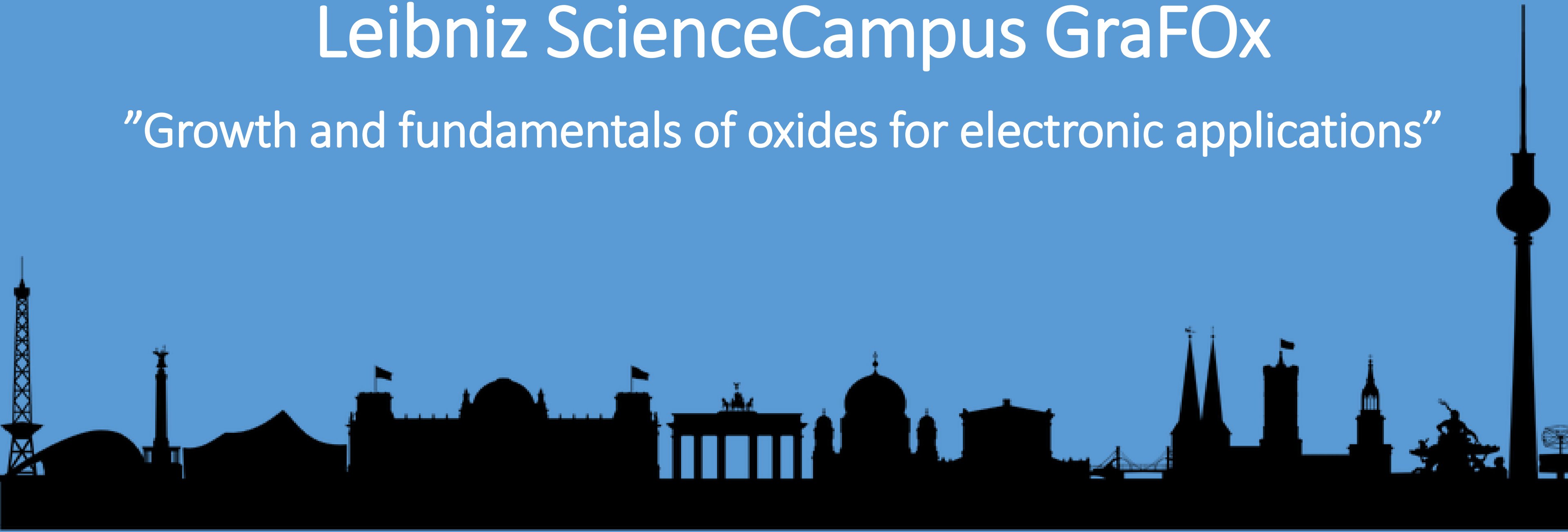
Andreas Popp/Günther Wagner,
Zbigniew Galazka

Leibniz GraFOx ScienceCampus

Leibniz Association

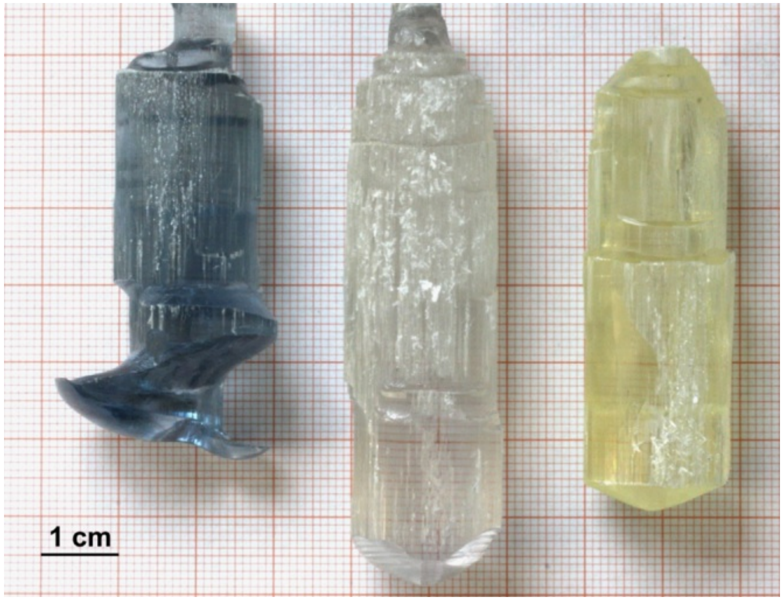
Leibniz ScienceCampus GraFOx

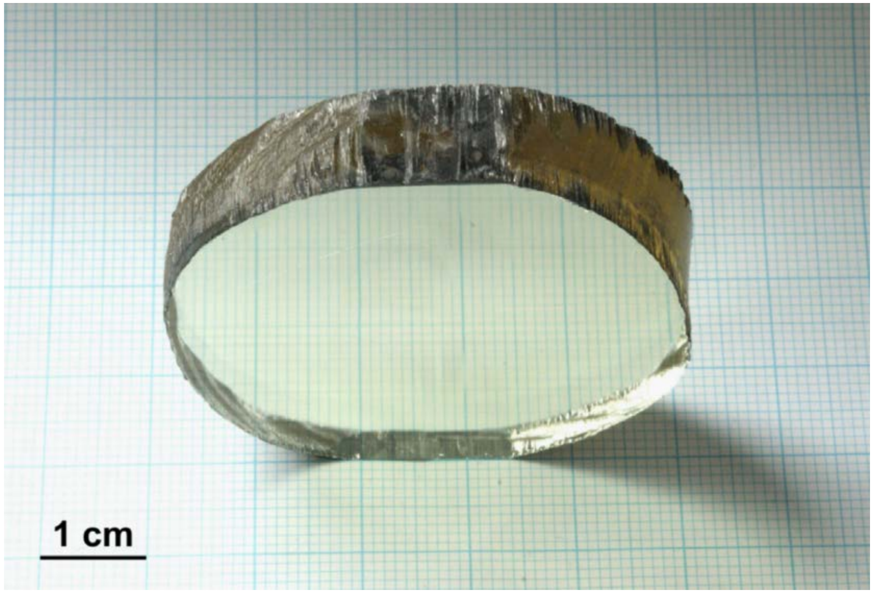
”Growth and fundamentals of oxides for electronic applications”



A transparent wide-band gap semiconductor

ß-Ga_2O_3





Galazka *et al.*,
Journal of Crystal Growth **404**, 184–191 (2014).

Single crystal growth: Z. Galazka

Leibniz Institute for Crystal Growth, Berlin, Germany

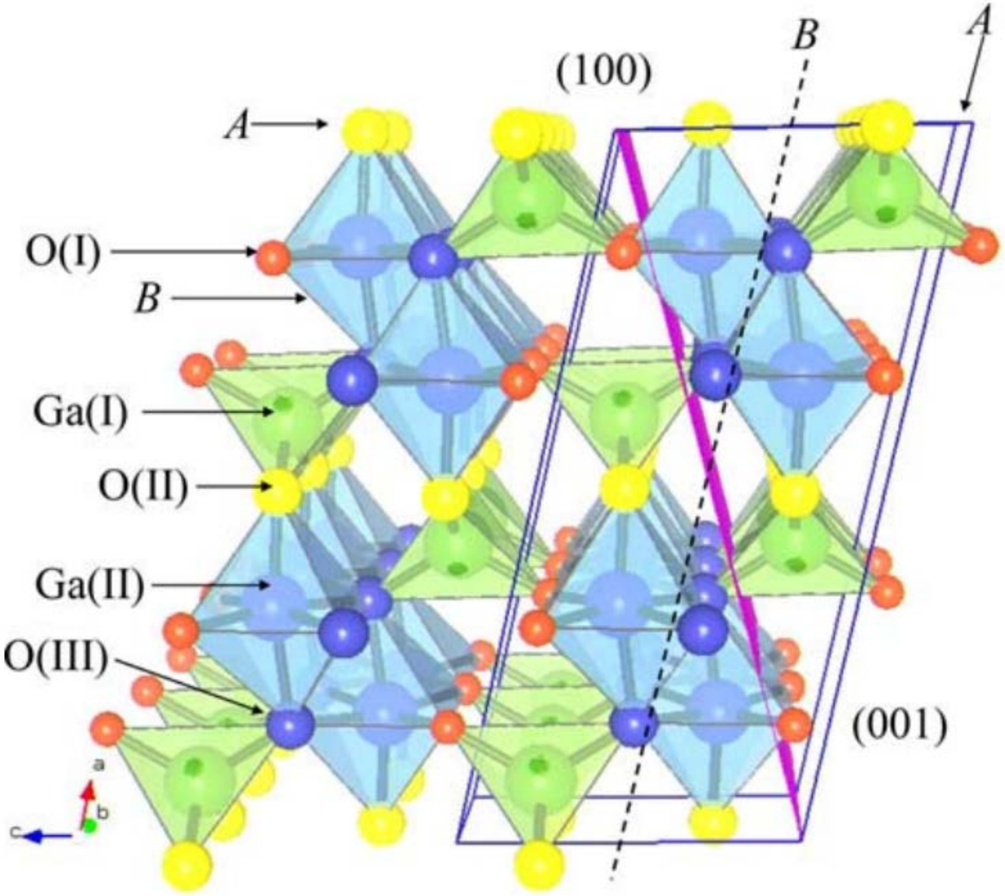
ß-Ga_2O_3 : Monoclinic crystal structure

$a = 12.214$ Å
$b = \ \ 3.037$ Å
$c = \ \ 5.798$ Å

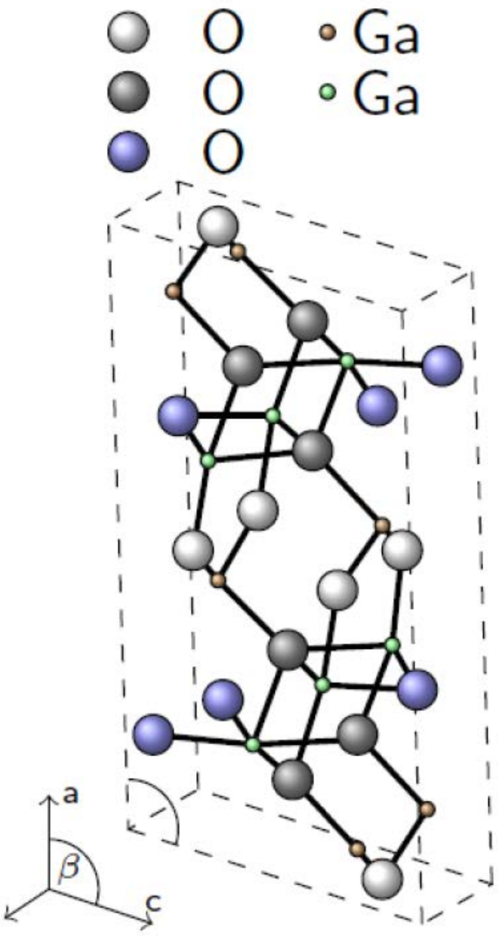
$$\boxed{\beta = 103.83°}$$

cleavage planes:

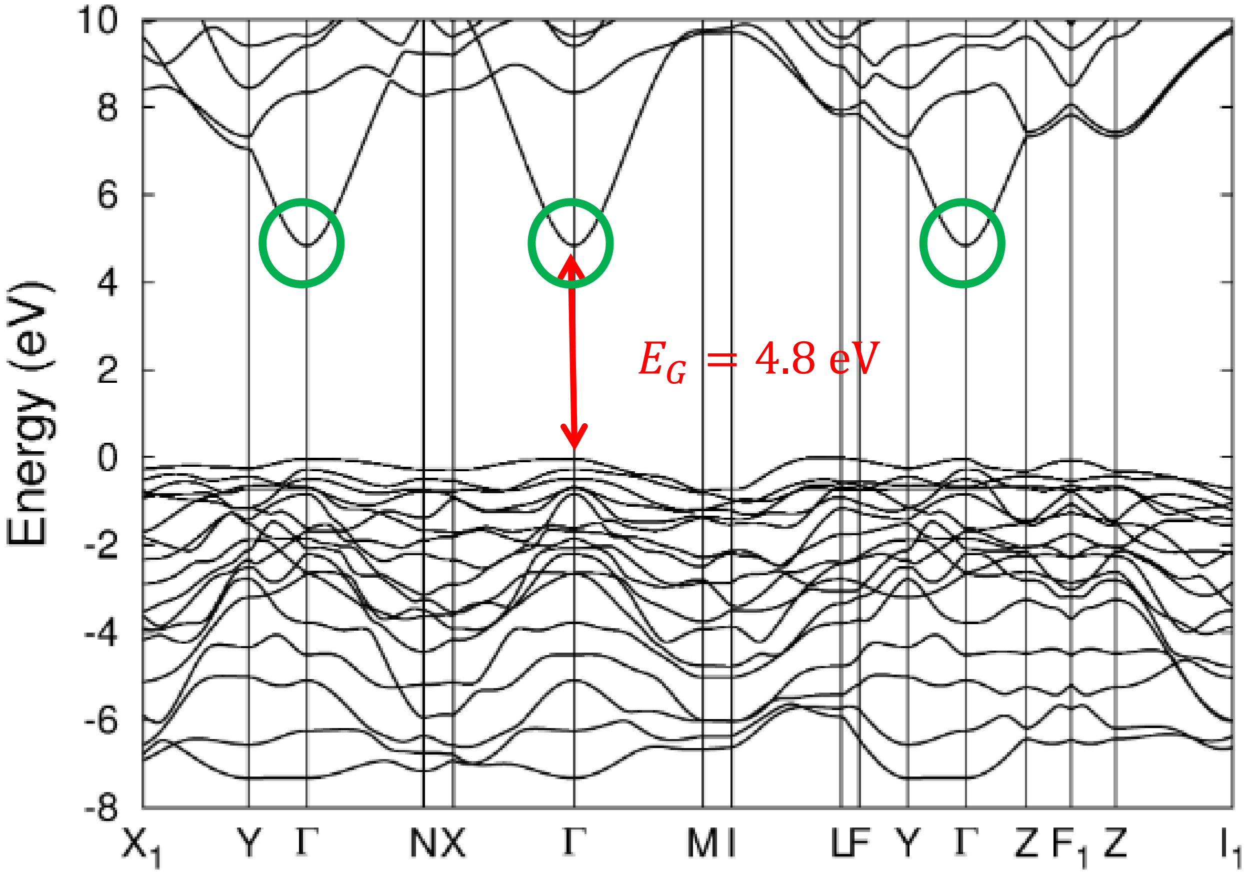
(100), (001)



H. Peelaers *et al.*, Phys. Status Solidi B **252**, No. 4 (2015)



ß-Ga_2O_3 : Electronic band structure



V. M. Bermudez, *Chem. Phys.* **323** 193 (2006)

Charge carriers: Electrons

Transport: parabolic band approx.

effective electron mass

$0.313\ m_e$

Mohamed, *et al.*, *Journal of Physics: Conference Series* **286**, 012027 (2011).

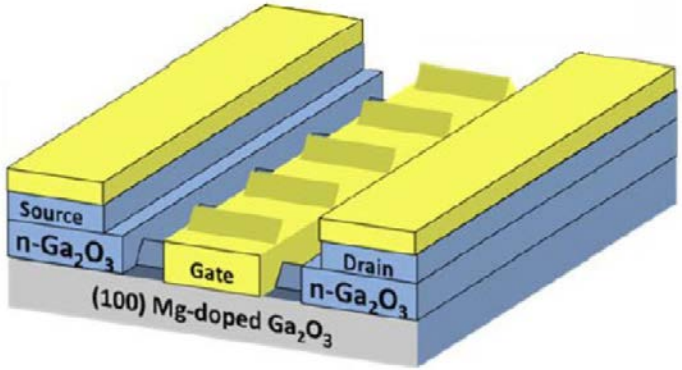
Opportunity of high breakdown field in Ga_2O_3

for high-power-devices

- E_{br} *predicted* to be ~ 8 MV/cm

  key advantage of Ga_2O_3

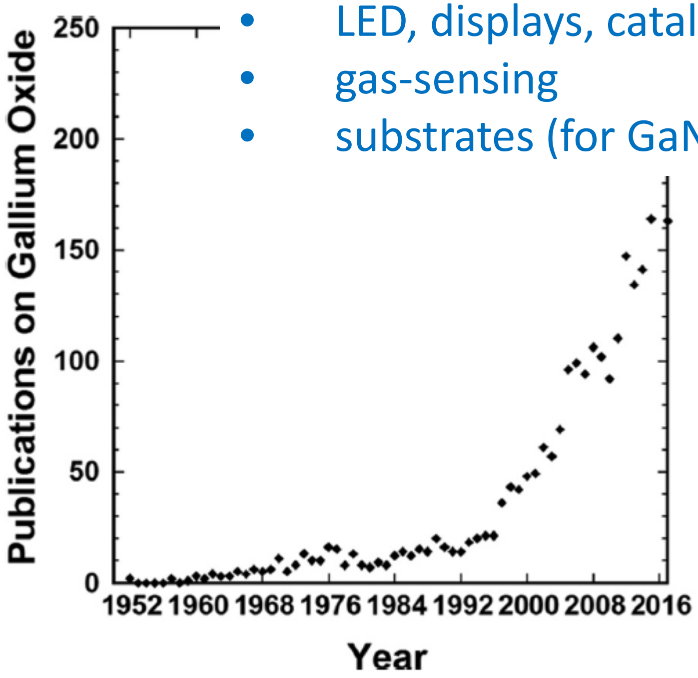
- Larger than the theoretical limits for GaN and SiC



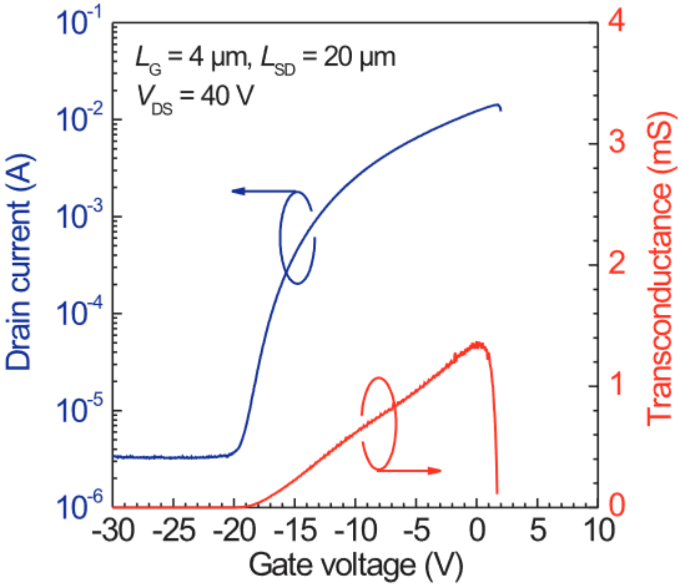
Chabak *et al.,*
Appl. Phys. Lett. **109**, 213501 (2016).

applications also in…

- LED, displays, catalysts
- gas-sensing
- substrates (for GaN)

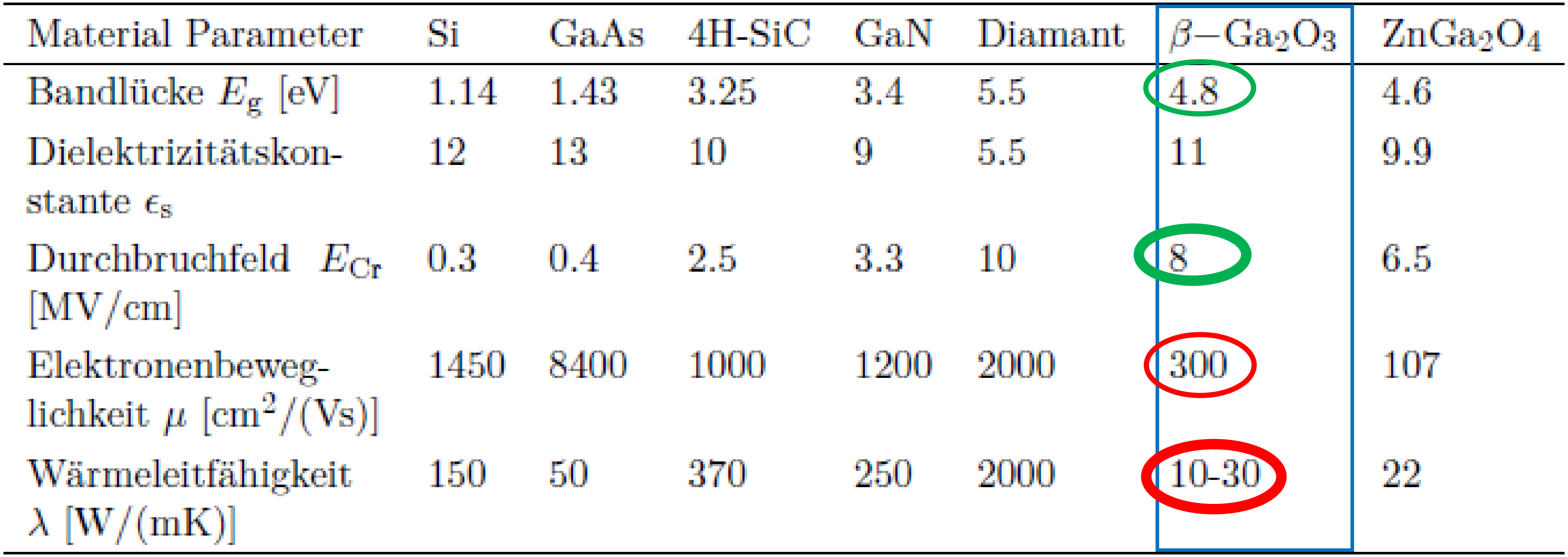


Pearton *et al.*, *Appl. Phys. Rev.* **5**, 011301 (2018).



M. Higashiwaki, *et al.;*
Appl. Phys. Lett. **100**, 013504 (2012).
Review (2022)

Material properties



Material Parameter	Si	GaAs	4H-SiC	GaN	Diamant	$\beta-\mathrm{Ga_2O_3}$	$\mathrm{ZnGa_2O_4}$
Bandlücke E_g [eV]	1.14	1.43	3.25	3.4	5.5	4.8	4.6
Dielektrizitätskonstante ϵ_s	12	13	10	9	5.5	11	9.9
Durchbruchfeld E_{Cr} [MV/cm]	0.3	0.4	2.5	3.3	10	8	6.5
Elektronenbeweglichkeit μ [cm^2/(Vs)]	1450	8400	1000	1200	2000	300	107
Wärmeleitfähigkeit λ [W/(mK)]	150	50	370	250	2000	10-30	22

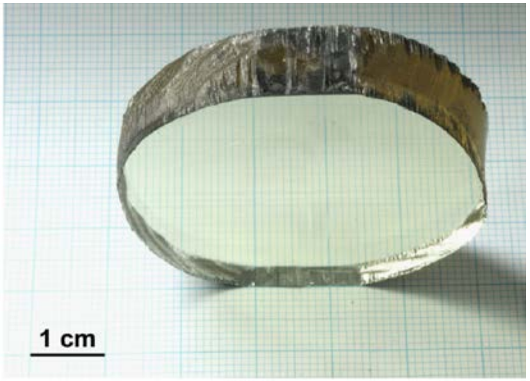
J. Boy, *PhD Thesis (2022)*

ß-Ga_2O_3-bulk single crystals and homoepitaxial films

Bulk single crystals:

Czochralski-growth

- As-grown, no doping
- $n = 9 \cdot 10^{16} - 6.5 \cdot 10^{17}\ \mathrm{cm}^{-3}$
- $d = 233 - 525\ \mu\mathrm{m}$



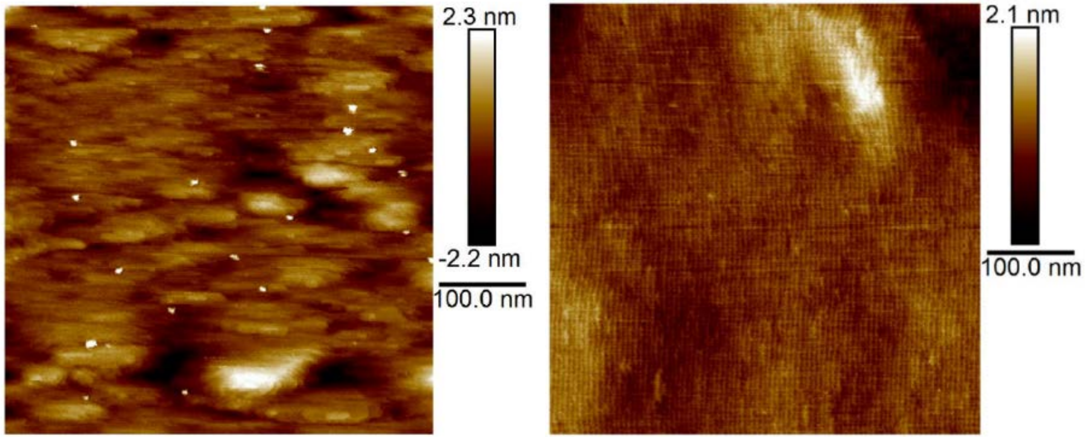
Galazka *et al.*,
J .Crystal Growth **404**,
184–191 (2014).

Z. Galazka, IKZ

Epitaxial layers:

Metallorganic Vapor Dep. (MOCVD)

- Si-Doping
- $n = 2.5 \cdot 10^{17} - 1.6 \cdot 10^{18}\ \mathrm{cm}^{-3}$
- $d = 25 - 225\ \mathrm{nm}$
- 2D island growth & (rough) step flow growth



Mohamed, *et al.*, *J. Physics: Conf. Series* **286**, 012027 (2011).

Groups of G. Wagner / A. Popp, IKZ

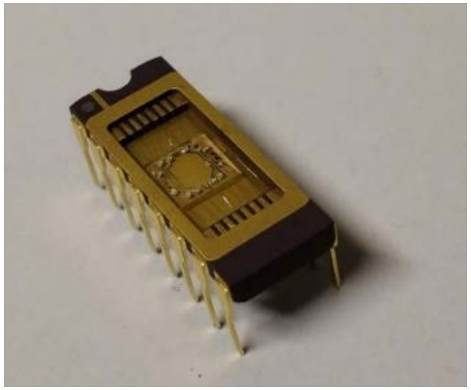
ß-Ga_2O_3 – A transparent wide-band gap *semiconductor*

- **Electrical conductivity**
- **Thermal conductivity**
- **Thermo-electricity**

bulk
&
thin film effects

Outlook: Giant-phonon drag *increase* by thin-film design

Thermo-/electric micro measurement platform for bulk & thin films



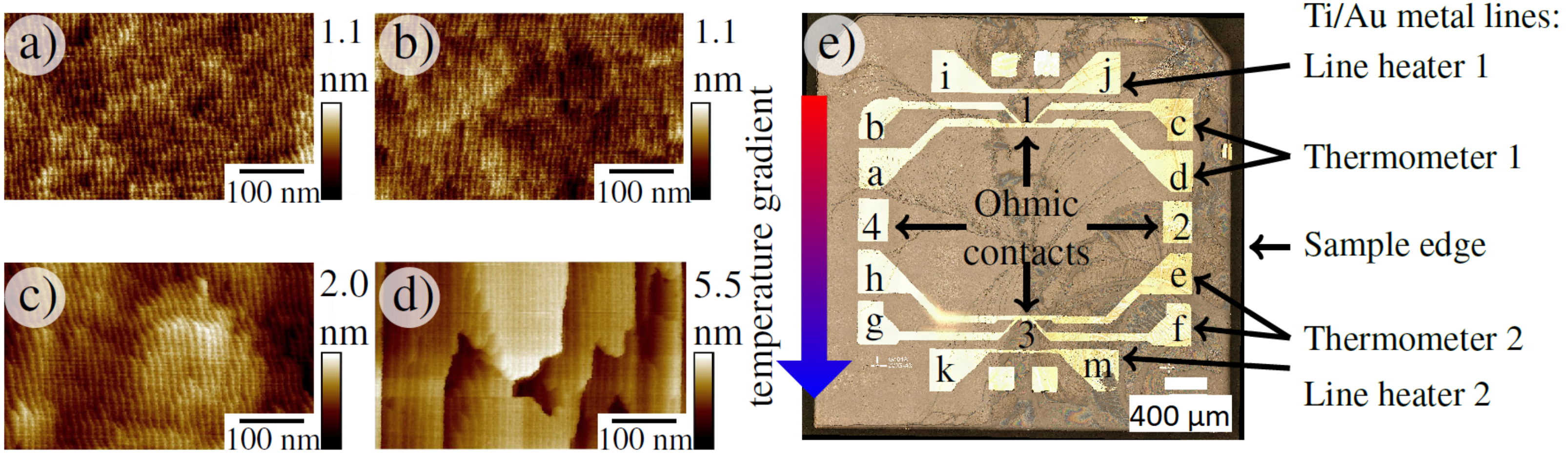


FIG. 1. (a) to (d) exemplary AFM measurement results of the investigated samples. (a) is the substrate of the $d = 152$ nm sample, (b) the substrate of the $d = 50$ nm sample, (c) the $d = 152$ nm thin film and (d) the $d = 50$ nm thin film. (e) Microscopic picture of a β-Ga_2O_3 thin film with the thermoelectric measurement platform consisting of Ti/Au (7 nm/35 nm) metal lines. Ohmic contacts were achieved by Al-wedge bonding at (1) - (4). The line heater and thermometer (a) - (m) were contacted by gold wires with indium contacts.

J. Boy, *et al.*, *APL Mater.* **7**, 022526 (2019).

Electrical properties

Bulk

Thick epitaxial films

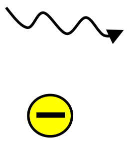
Thin epitaxial films

➔ Limiting effects of electron mobility
- even vor ideal thin films

A „zoo" of scattering mechanisms

phonon

electron



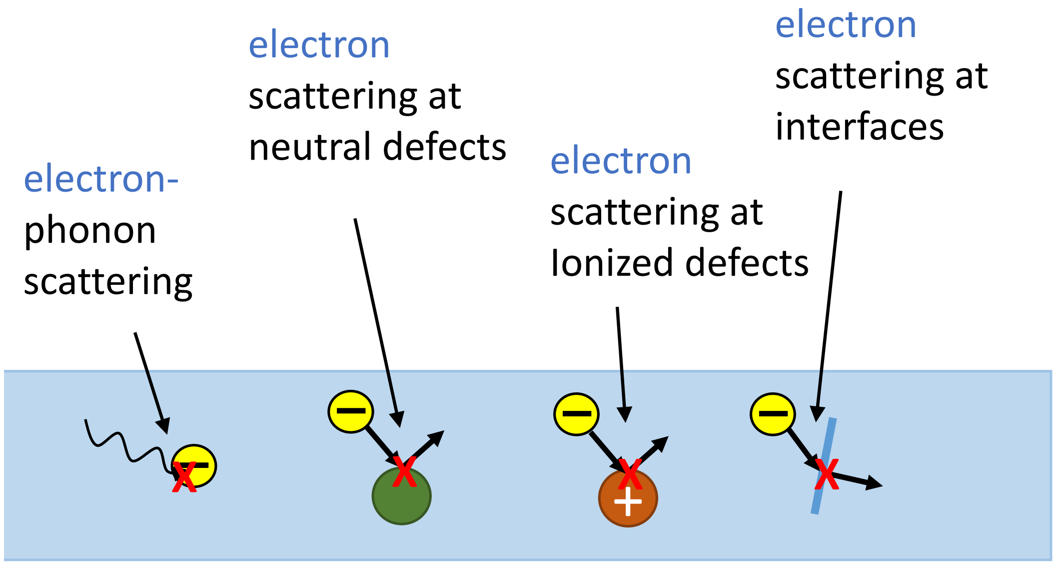
$$\sigma = \frac{e^2 n}{m^*}\langle\tau\rangle, \quad R_{\mathrm{H}} = \frac{1}{ne}\frac{\langle\tau^2\rangle}{\langle\tau\rangle^2},$$

Reduced by…

Temperature

Expitaxy

Homoepitaxy



electron-phonon scattering

electron scattering at neutral defects

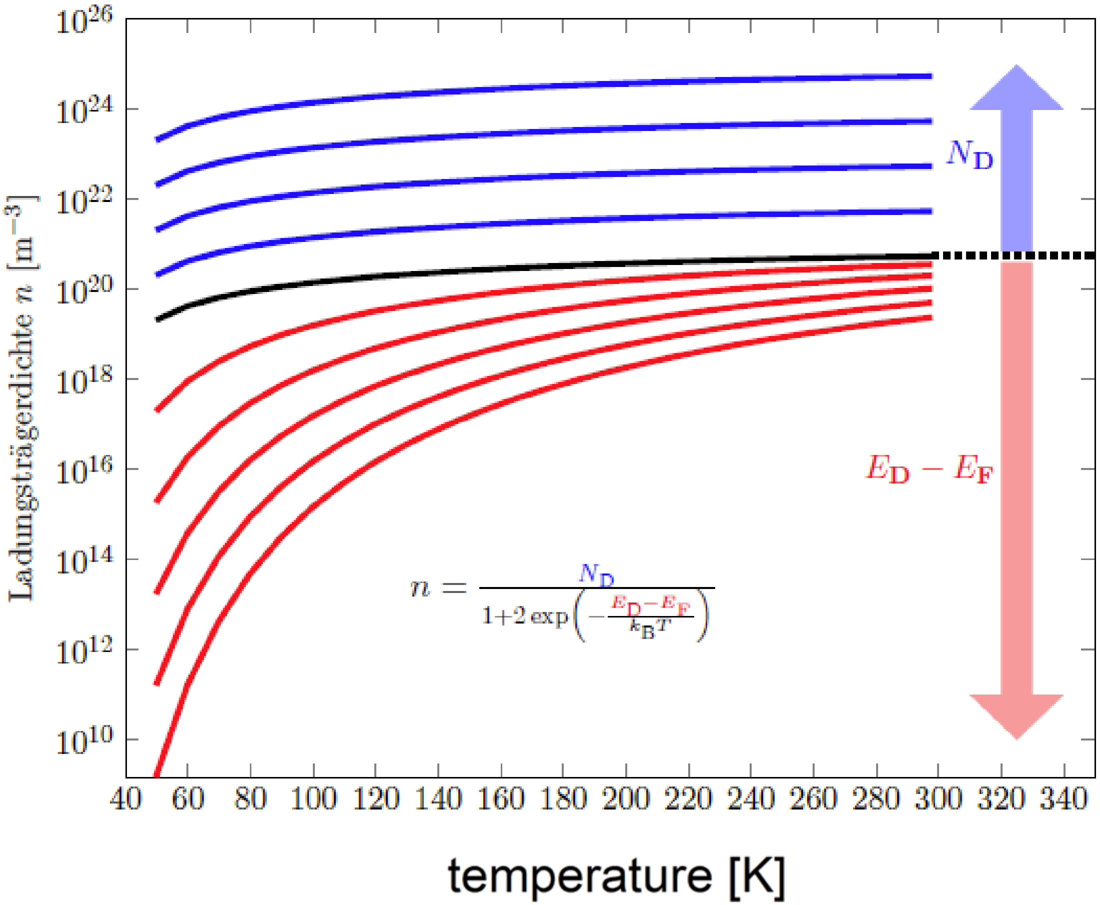
electron scattering at Ionized defects

electron scattering at interfaces

Electrical properties

Electron density

$$n = \int_0^\infty D(E) f(T,E) \mathrm{d}E \qquad n = \int_0^\infty \frac{(2m^*)^{\frac{3}{2}}}{2\pi^2\hbar^3} \cdot \frac{\sqrt{E+E_\mathrm{F}-E_\mathrm{C}}}{1+\exp\left(\frac{E}{k_\mathrm{B}T}\right)} \mathrm{d}E$$



T. Oishi, *et al.*;
Appl. Phys. Express **8**, 031101 (2015).

J. Boy, *PhD Thesis (2022)*

Electrical properties

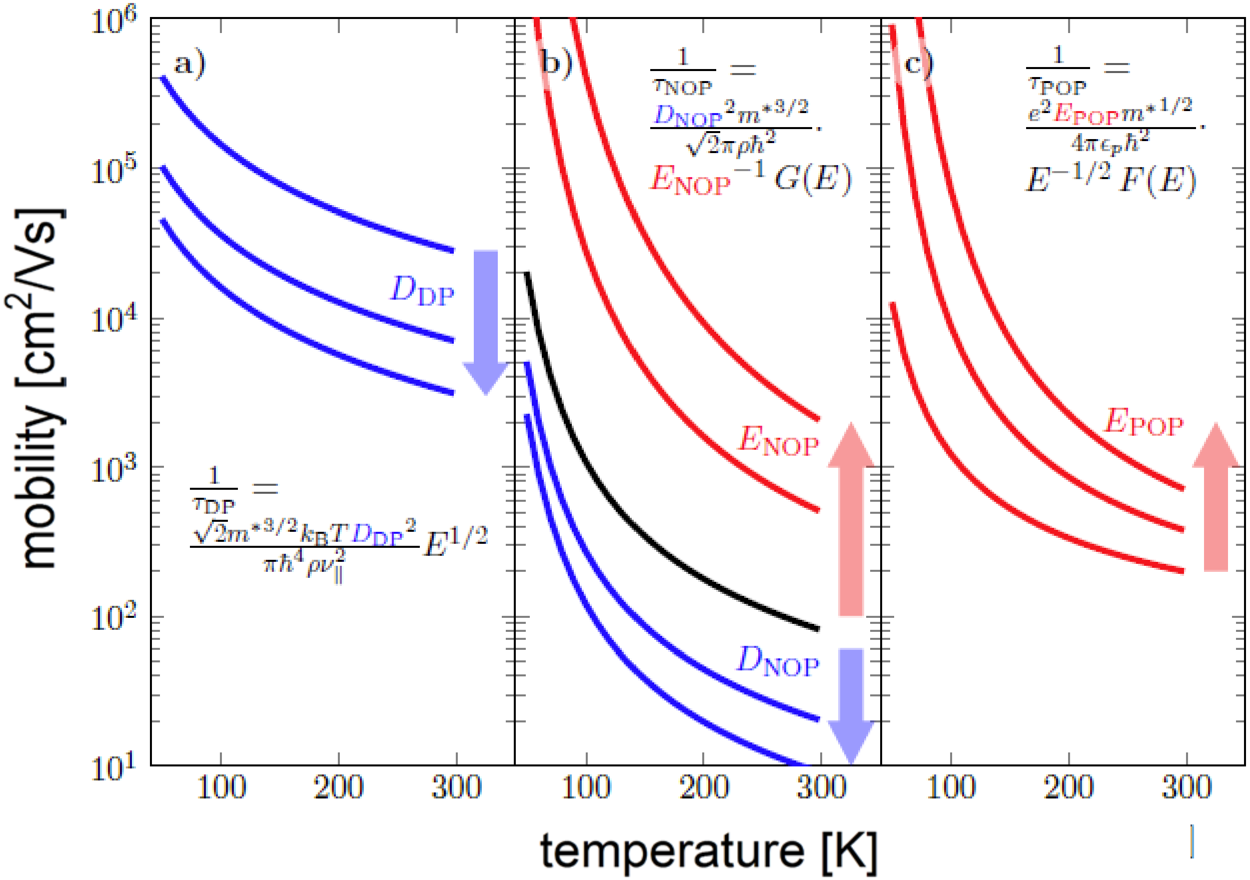
Electron mobility

$$\mu = \frac{e\langle\tau_m\rangle}{m^*} = \frac{e}{m^*}\frac{\int_0^\infty E^{3/2}\tau_m(E)f(E)\mathrm{d}E}{\int_0^\infty E^{3/2}f(E)\mathrm{d}E}.$$

acoustical phonons

non-polar optical phonons

polar optical phonons



scattering by phonons

J. Boy, *PhD Thesis (2022)*

Electrical properties

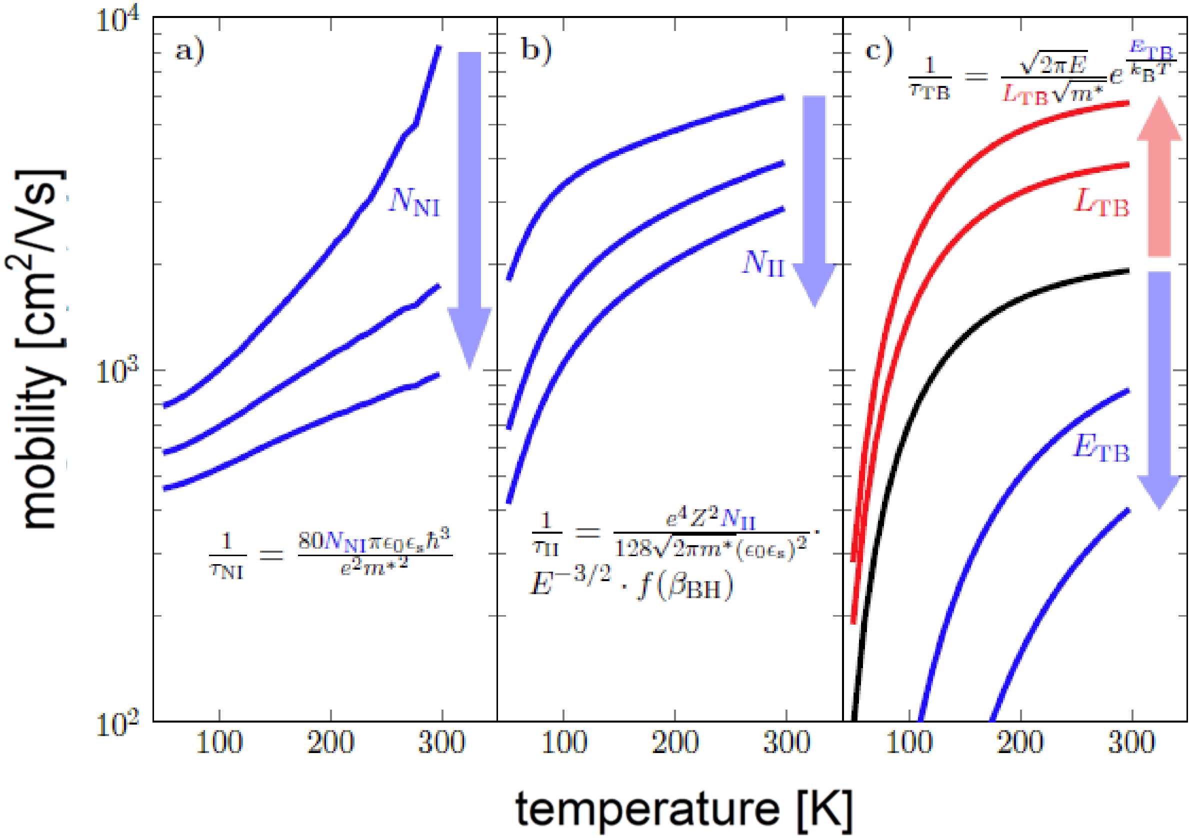
Electron mobility

$$\mu = \frac{e\langle\tau_m\rangle}{m^*} = \frac{e}{m^*}\frac{\int_0^\infty E^{3/2}\tau_m(E)f(E)\mathrm{d}E}{\int_0^\infty E^{3/2}f(E)\mathrm{d}E}.$$

neutral defects

ionized defects

planar defects

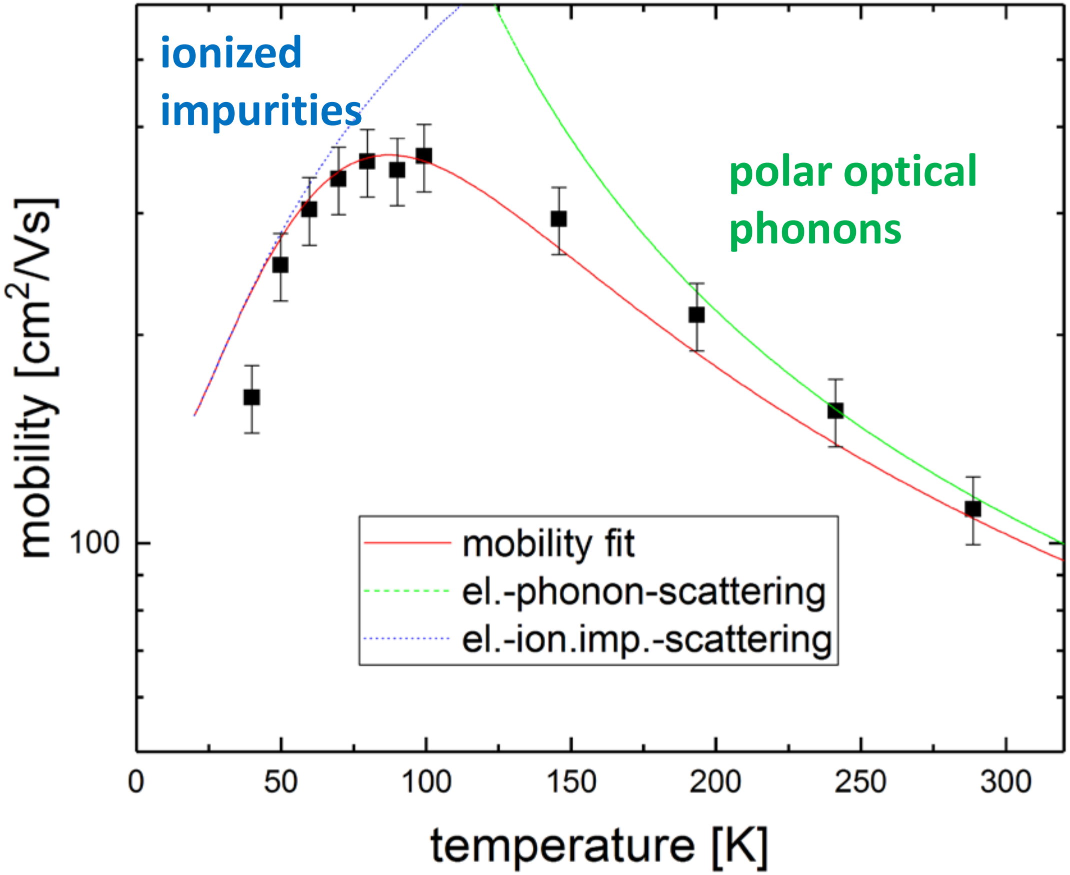


scattering by defects

J. Boy, *PhD Thesis (2022)*

Charge transport in homoepitaxial ß-Ga_2O_3-bulk

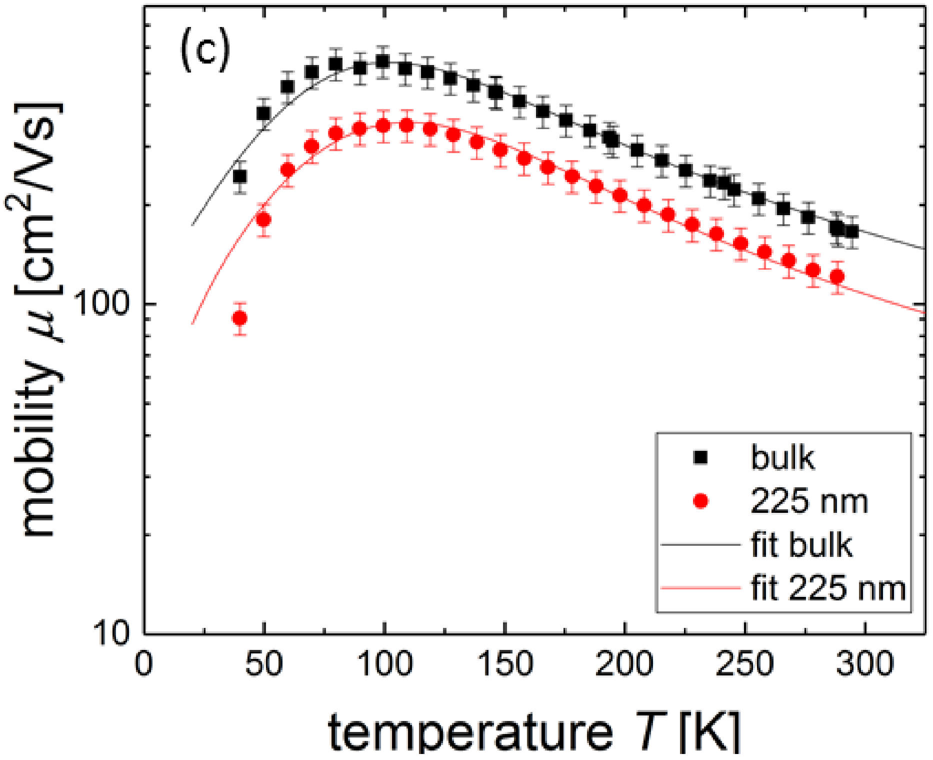
Dominant scattering mechanisms in ß-Ga_2O_3 single crystal



R. Ahrling, *et al.*; Scientific Reports **9,** 13149 (2019).

Charge transport in homoepitaxial ß-Ga_2O_3-films

single crystal bulk & 200 nm film

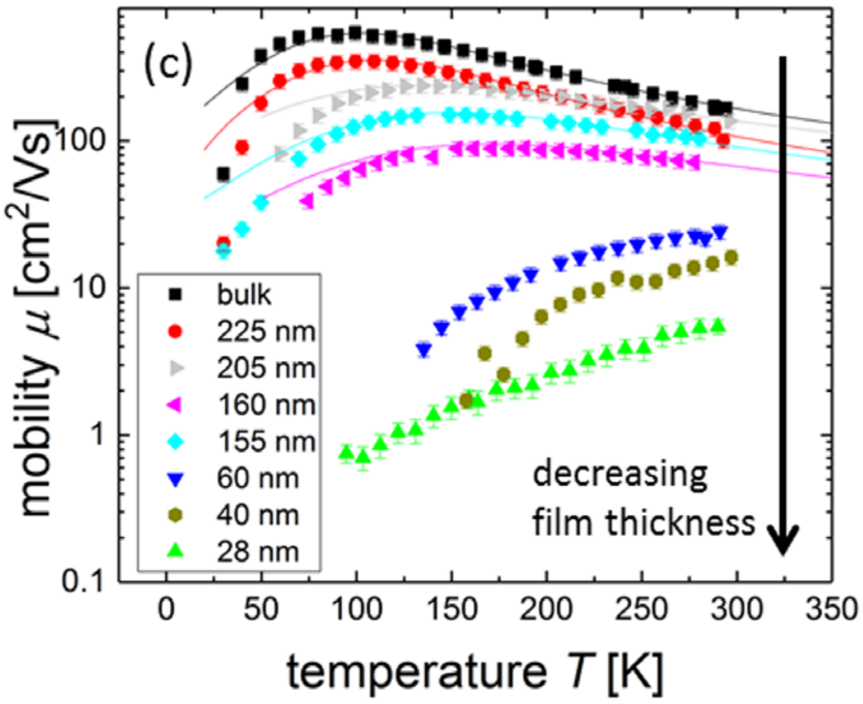


➔ **high-quality homoepitaxial films**

R. Ahrling, *et al.*; Scientific Reports **9,** 13149 (2019).

Charge transport in homoepitaxial ß-Ga_2O_3-films

Mobility suppression with decreasing film thickness



Real films:

- Neutral impurities, hopping transport
- **Twin boundary scattering**

$$\mu_{\text{tb}} = \frac{eL}{\sqrt{8k_B T \pi m^*}} \exp\left(-\frac{E_B}{k_B T}\right)$$

J.W. Orton, *et al.*;
Rep. on Prog. in Physics **43**, 1263 (1980).
R. Schewski, *et al.*;
J. of Appl. Phys. **120**, 225308 (2016).

- **… but also for ideal thin films:**

➔ Surface scattering
&
boundary effects

R. Ahrling, *et al.*; Scientific Reports **9,** 13149 (2019).

Charge transport in homoepitaxial ß-Ga_2O_3-films

Surface scattering & boundary effects

- **length scales:**

thickness t
mean free path l
de Broglie wavelength λ_e
surface roughness r_S

- **In metals:** $\boldsymbol{l \approx t\,, \lambda_e \ll l, r_S \gg \lambda_e}$
  - Fuchs-Sondheimer model: l/t determines mobility

K. Fuchs, Math. Proc. of the Camebridge Philosophical Society **34**, 100 (1938).
E. Sondheimer, Advances in Physics **1**, 1 (1952).

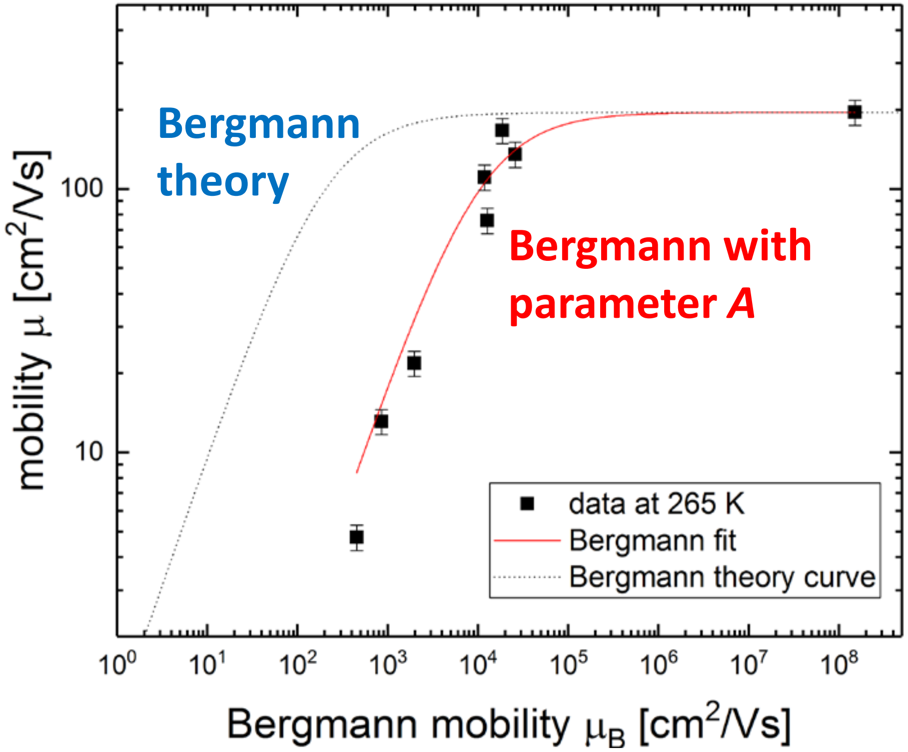
- **Here:** $\boldsymbol{\lambda_e \approx t \quad , \quad \lambda_e \gg l \quad , \quad \lambda_e \gg r_S}$
  - Bergmann model: quantum mechanical waveguide effect

$$\boxed{\mu_{\text{Bergmann}} = \frac{e}{\hbar}\left(\frac{t}{\lambda_e}\right)^2 \ln\left(\frac{t}{\lambda_e}\right)\frac{1}{nt}}$$

G. Bergmann, *et al.*; PRL **94**, 106801 (2005).

Charge transport in *ideal* thin films

Bergman model



$$\mu_{\text{tot}} = \left(\frac{1}{A \cdot \mu_{\text{Bergmann}}} + \frac{1}{\mu_{\text{vol}}} \right)^{-1}$$

- Mobilities fit to Bergmann model

- Quantitative agreement for $A = 0.02$

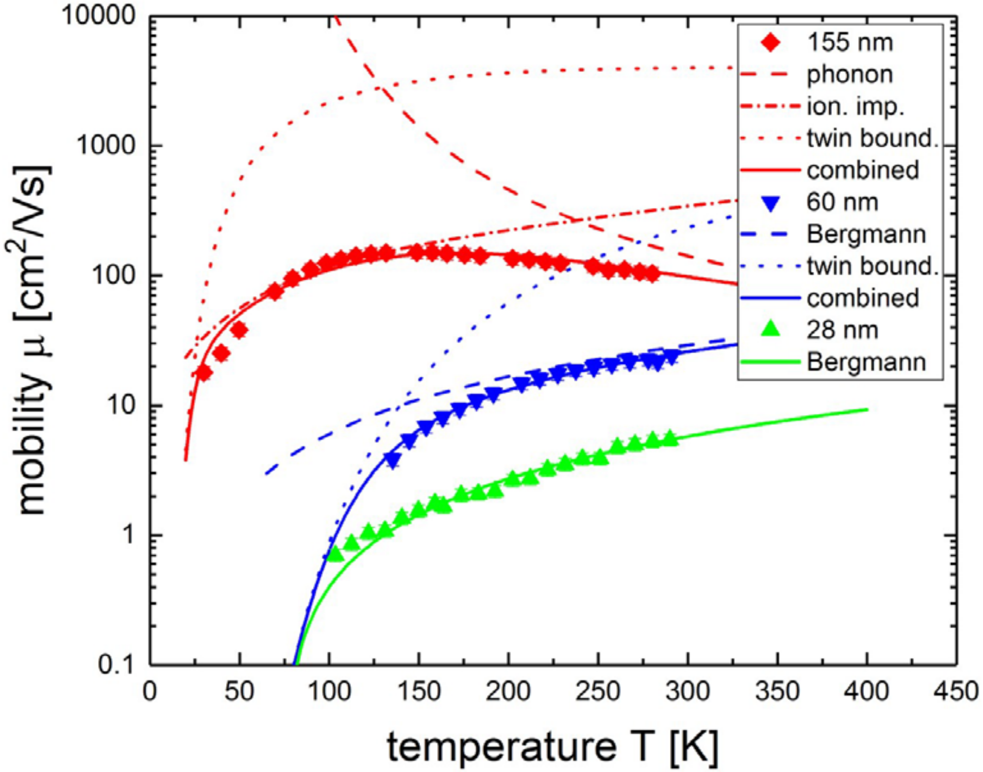
G. Bergmann, *et al.*,
PRL **94**, 106801 (2005).

R. Ahrling, *et al.*; Scientific Reports **9,** 13149 (2019).

Charge transport in homoepitaxial ß-Ga_2O_3-films

Summary

- **Thick homoepitaxial (100) β-Ga_2O_3 films (above 150 nm) behave bulk-like**
  - Optical phonon scattering dominates μ for high T
  - Ionized impurity scattering dominates μ for low T
- **Thin films (below 100 nm) decrease in and change in $\mu(T)$ behavior**
  - Additional scattering mechanism occurs → mobility reduction
  - Ideal films: Described by quantum mechanical waveguide effect
- Mobility reduction has to be taken into account for use of thin β-Ga_2O_3 films in devices



R. Ahrling, *et al.*; Scientific Reports **9,** 13149 (2019).

Thermal properties

Bulk

→ Anisotropy in thermal conductivity

Homoepitaxial films

→ Phonon-transparent interfaces

Ballistic phonon transport

Thermal transport measurements

Thermal conduction differential equation:

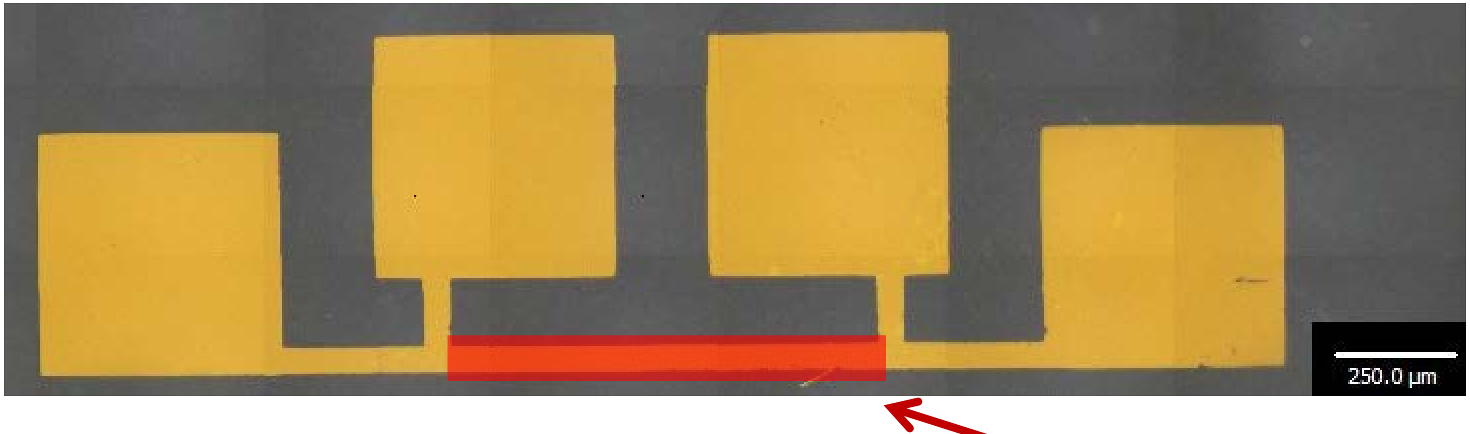
$$\frac{\partial^2 \Delta T(r,t)}{\partial r^2} + \frac{1}{D}\frac{\partial \Delta T(r,t)}{\partial t} = 0$$

thermal diffusivity: $\widehat{D}$

thermal conductivity: $\hat{\lambda} = \widehat{D} \cdot C_V \cdot \rho$

D. G. Cahill, *et al.;*
Phys. Rev. B, **50**, 6077 (1994).

Experimental setup – electrical line heater



Solution:

$$\Delta T(r) = \frac{P}{2\pi\hat{\lambda}L} K_0(qr)$$

heating power

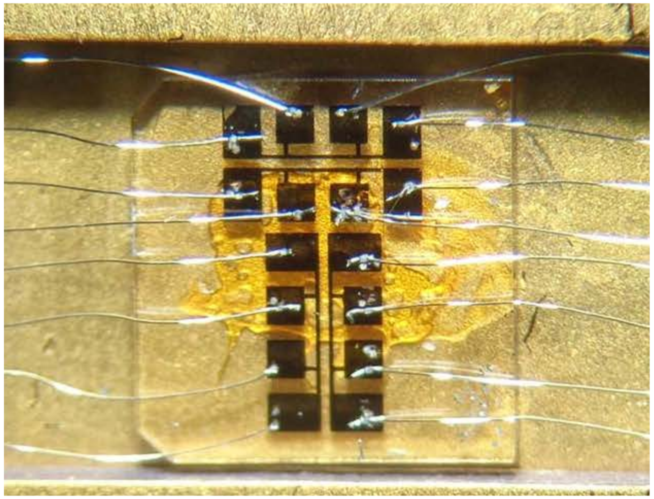
Bessel function (zero order second kind)

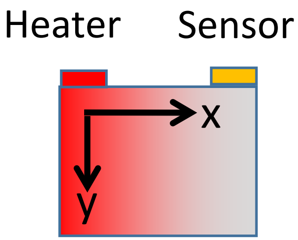
inverse thermal penetration depth

$q = \sqrt{i2\omega/\widehat{D}}$

Thermal transport measurements

2ω-method for anisotropy characterization

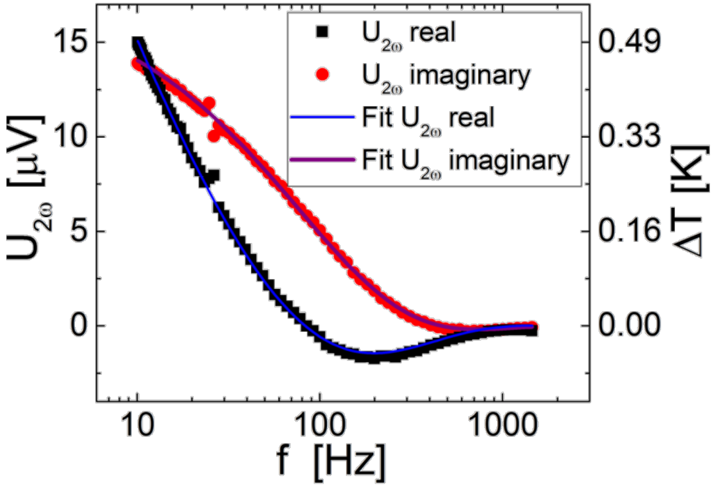




A. T. Ramu and J. E. Bowers;
Rev. Sci. Instr. **83** 124903 (2012).

$$\Delta T = \frac{P}{\pi L \bar{\lambda}} \frac{1}{2\omega_\mathrm{h}} \int_{-\omega_\mathrm{h}}^{\omega_\mathrm{h}} \frac{1}{2\omega_\mathrm{s}} \int_{-\omega_\mathrm{s}}^{\omega_\mathrm{s}} K_0\big(q \cdot (d + o - p)\big) \mathrm{d}o\mathrm{d}p$$
$$\propto U_{2\omega}$$

$$\bar{\lambda} = \sqrt{\lambda_x \cdot \lambda_y}$$
$$q = \sqrt{i 2\omega / D_x}$$

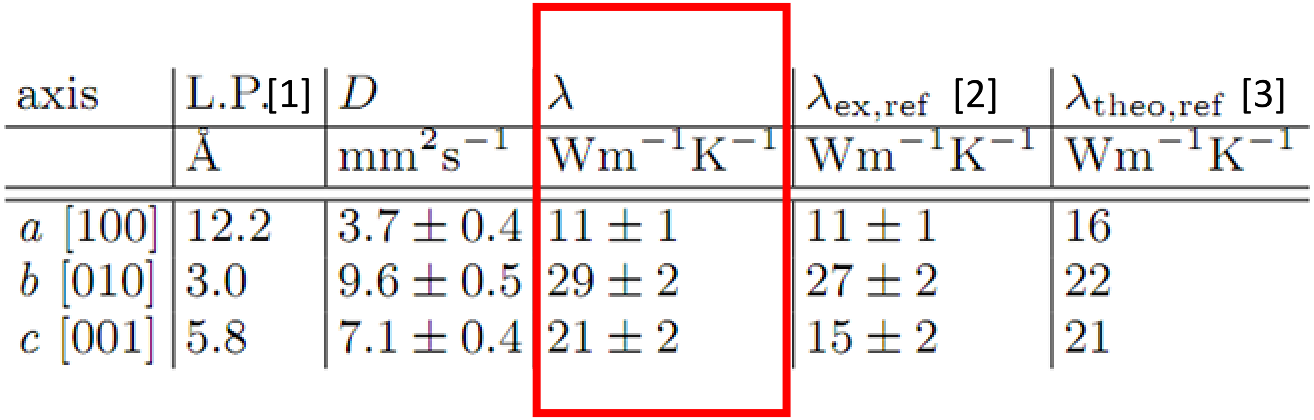


We obtain: $\bar{\lambda}_{[100],[001]}$, $\bar{\lambda}_{[100],[010]}$, $D_{[001]}$, $D_{[010]}$

Thermal transport in ß-Ga_2O_3 single crystals

Room temperature: thermal diffusivity D and thermal conductivity λ for bulk

- [100]-oriented Czochralski grown insulating Mg-doped **β-Ga_2O_3** single-crystal
- Diffusivity D and conductivity λ: $\boxed{\lambda = D \cdot C_V \cdot \rho}$



axis	L.P.[1]	D	λ	$\lambda_{\mathrm{ex,ref}}$ [2]	$\lambda_{\mathrm{theo,ref}}$ [3]
	Å	$\mathrm{mm^2s^{-1}}$	$\mathrm{Wm^{-1}K^{-1}}$	$\mathrm{Wm^{-1}K^{-1}}$	$\mathrm{Wm^{-1}K^{-1}}$
a [100]	12.2	3.7 ± 0.4	11 ± 1	11 ± 1	16
b [010]	3.0	9.6 ± 0.5	29 ± 2	27 ± 2	22
c [001]	5.8	7.1 ± 0.4	21 ± 2	15 ± 2	21

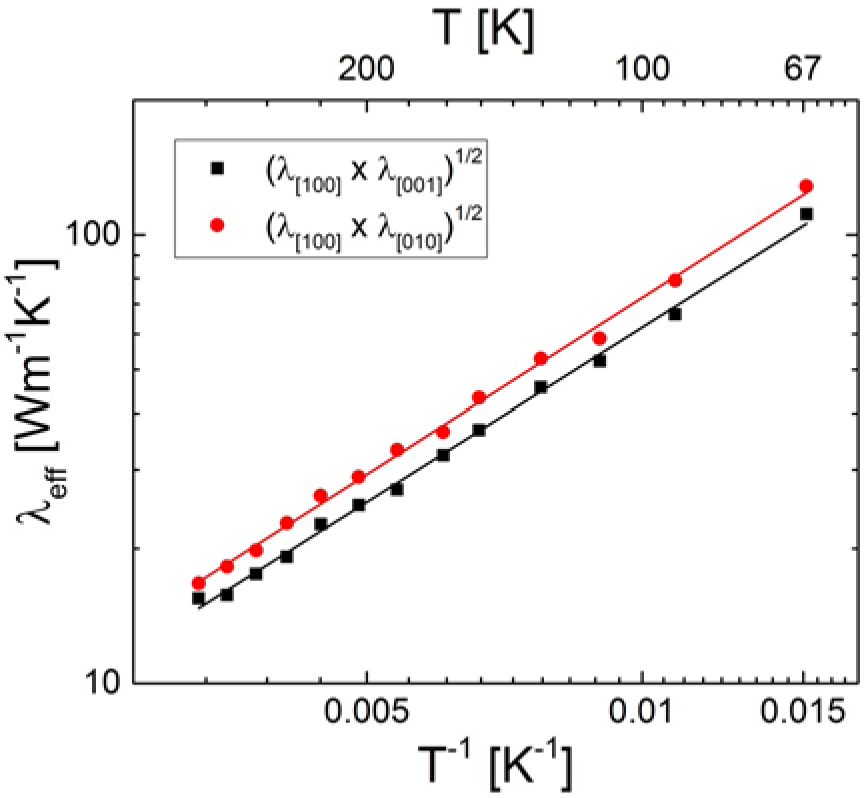
[1] V. M. Bermudez, *Chem. Phys.* **323** 193 (2006)
[2] Z. Guo *et al.* , Appl. Phys. Lett. **106**, 111909 (2015)
[3] M. D. Santia *et al.* Appl. Phys. Lett. **107**, 041907 (2015)

- **Highest thermal conductivity value along [010] –> (010) no cleavage plane**
- **Lowest thermal conductivity value along [100] –> (100) cleavage plane**

Thermal transport in ß-Ga_2O_3 single crystals

Temperature dependent thermal conductivity λ

- temperature-independent anisotropy factor: $\frac{\lambda_{[010]}}{\lambda_{[001]}} = 1.4 \pm 0.1$



M. Handwerg, *et al.;*
Semicond. Sci. Technol. **31**, 125006 (2016).

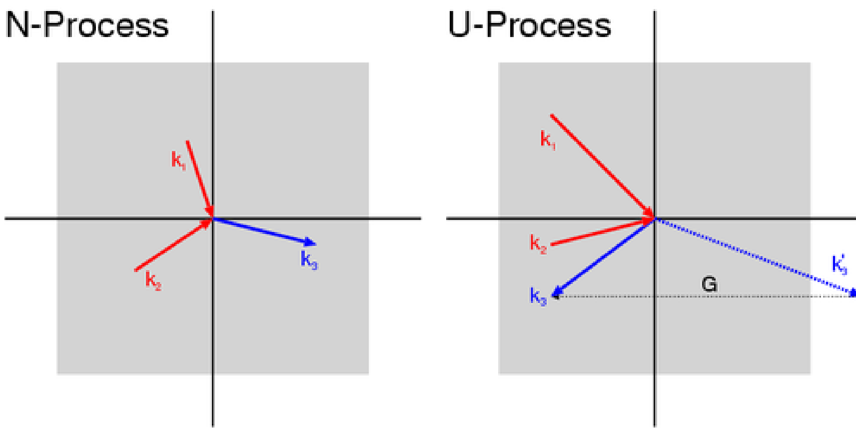
Temperature dependence:

$$\boxed{\lambda = \frac{1}{3} C_V(T)\Lambda(T)v_s}$$

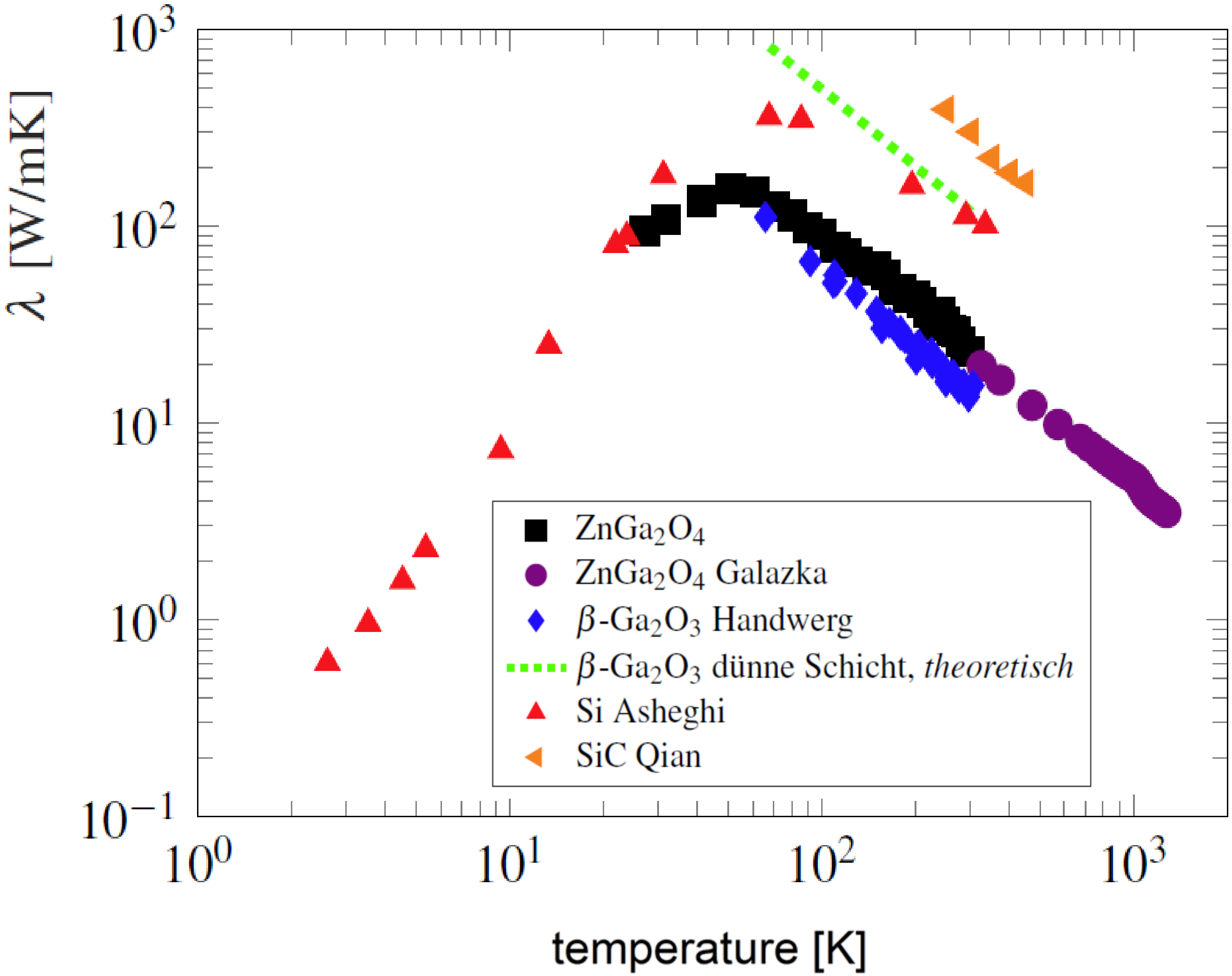
Solid line:

$C_V \cdot \Lambda \propto T^m$ with $m = 1.3 \pm\ 0.1$

phonon-phonon-Umklapp-scattering:



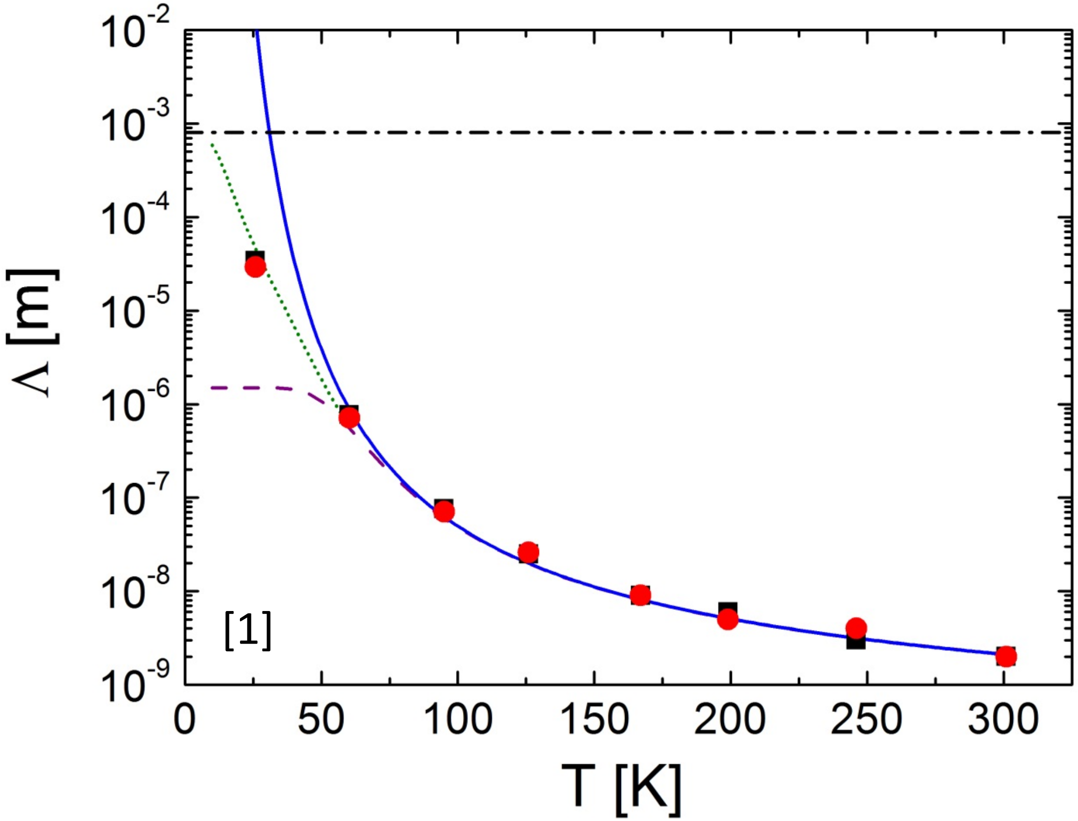
Comparision: Thermal conductivity λ



J. Boy, *PhD thesis (2023)*

Thermal transport in ß-Ga_2O_3 single crystals

Phonon mean free path



- For low temperatures or thin films the phonon mean free path reaches the sample size:

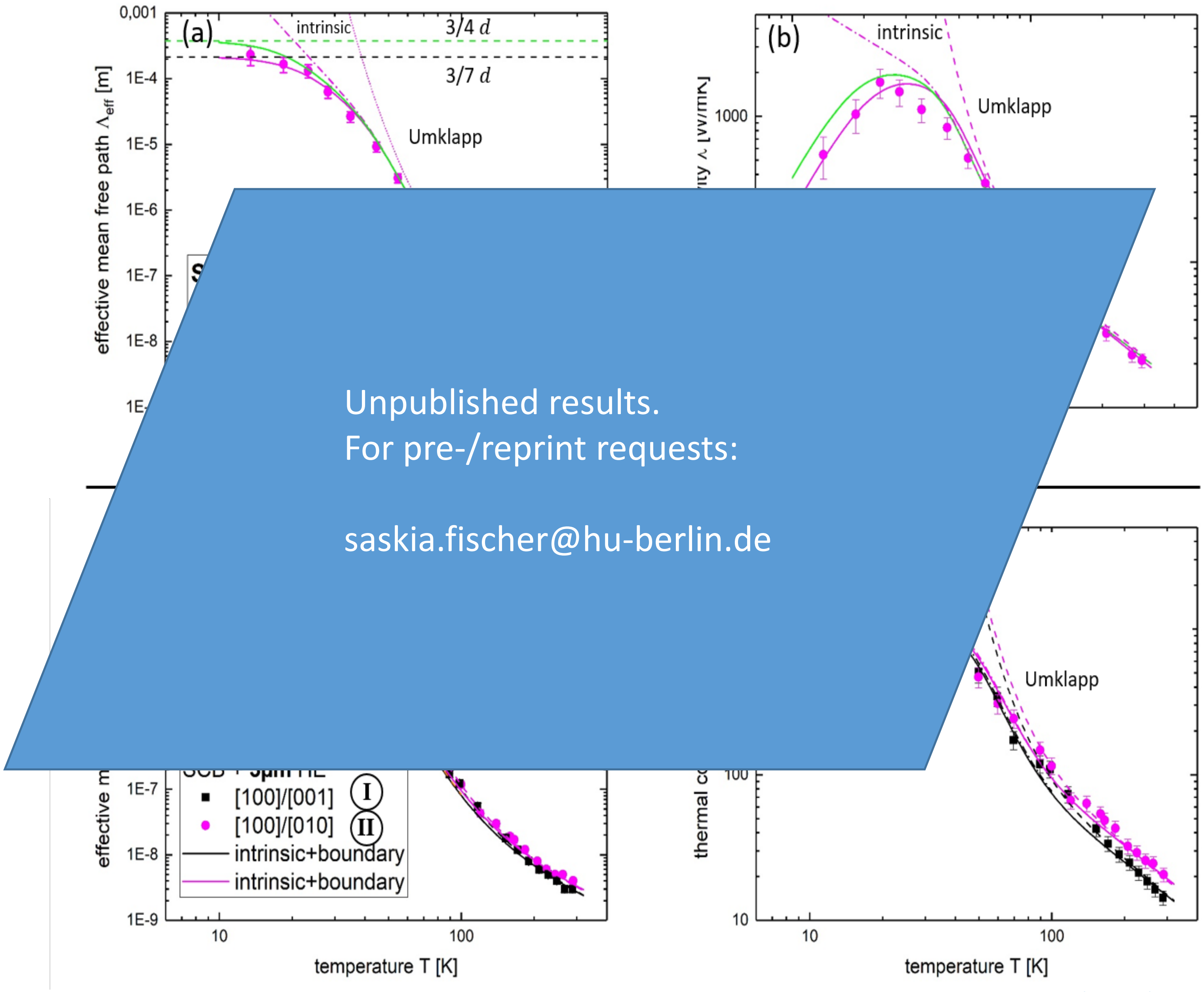
➔ **towards Casimir-limit**

- There the thermal conductivity is maximized

➔ **ballistic phonon transport**

M. Handwerg, *et al.;* Semicond. Sci. Techn. **30**, 024006 (2015).

Thermal transport in ß-Ga_2O_3 single crystals + homoepitaxial films



Unpublished results.
For pre-/reprint requests:

saskia.fischer@hu-berlin.de

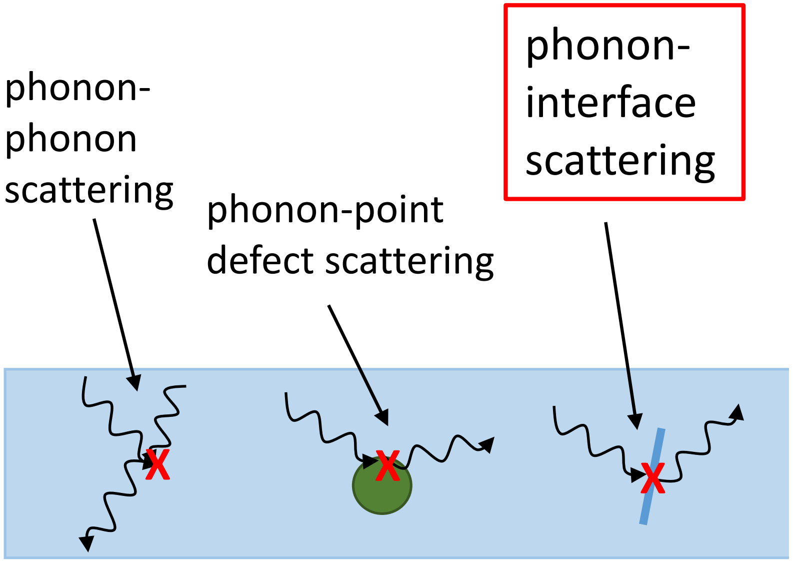
bulk

bulk
+
homoepi
film

R. Ahrling, *PhD thesis (2023)*

Phonon scattering mechanisms

phonon



Reduced by…

Temperature

Expitaxy

Homoepitaxy

Thermal transport in thin films

Summary

Mg-doped insulating **β-Ga_2O_3** bulk crystals and homo-epi films

- RT: $\boxed{\lambda_{[100]} = 11 \pm 1,}$ $\boxed{\lambda_{[010]} = 29 \pm 2}$ and $\boxed{\lambda_{[001]} = 21 \pm 2}$ W/(mK)
- Phonon-transparent interface in homoepitaxial films
- Ballistic phonon transport at low temperatures

A remark on polycrystalline films…

- thermal conductivity is decreased due to a reduced phonon mean free path.

M. Handwerg, *et al.;* Semicond. Sci. Techn. **30**, 024006 (2015).
M. Handwerg, *et al.;* Semicond. Sci. Techn. **31**, 125006 (2016).
R. Mitdank, *et al.;* Phys. Stat. Sol., A **211**, 543-549 (2014).
R. Ahrling, PhD Thesis (2023)

Thermoelectric properties

Bulk and thick epitaxial films

→ Diffusive TE & phonon drag

Thin epitaxial films

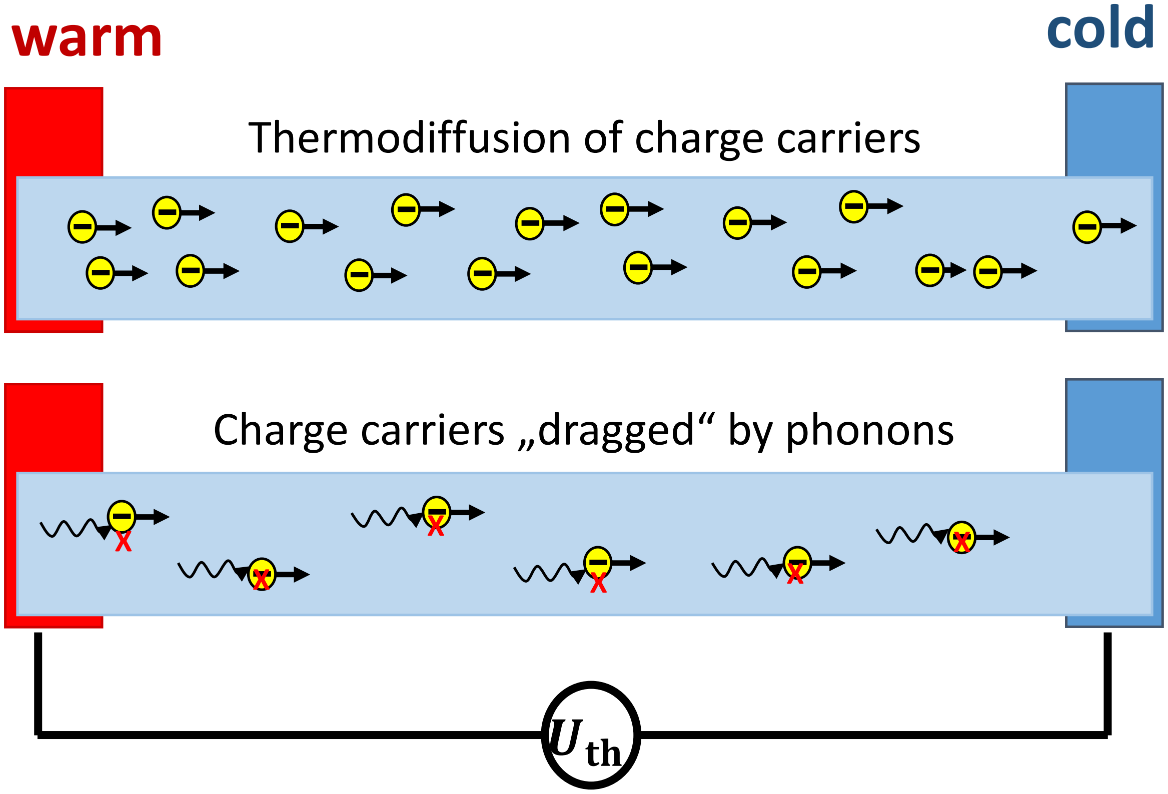
→ Phonon-transparent interfaces

Giant-phonon drag by design

Thermoelectric effects in semiconductors

Thermal gradient: ΔT

Electric field occurs by two processes:



warm

cold

Thermodiffusion of charge carriers

Charge carriers „dragged“ by phonons

U_{th}

Thermovoltage

C. Herring,
Phy. Rev. **96**, 1163 (1954).

Seebeck coefficient:

$$S = -\frac{U_{\mathrm{th}}}{\Delta T}$$

Thermoelectric effects in semiconductors

Seebeck coefficient:

$$\boldsymbol{S = -\frac{U_{\mathrm{th}}}{\Delta T} = S_{\mathrm{d}} + S_{\mathrm{PD}}}$$

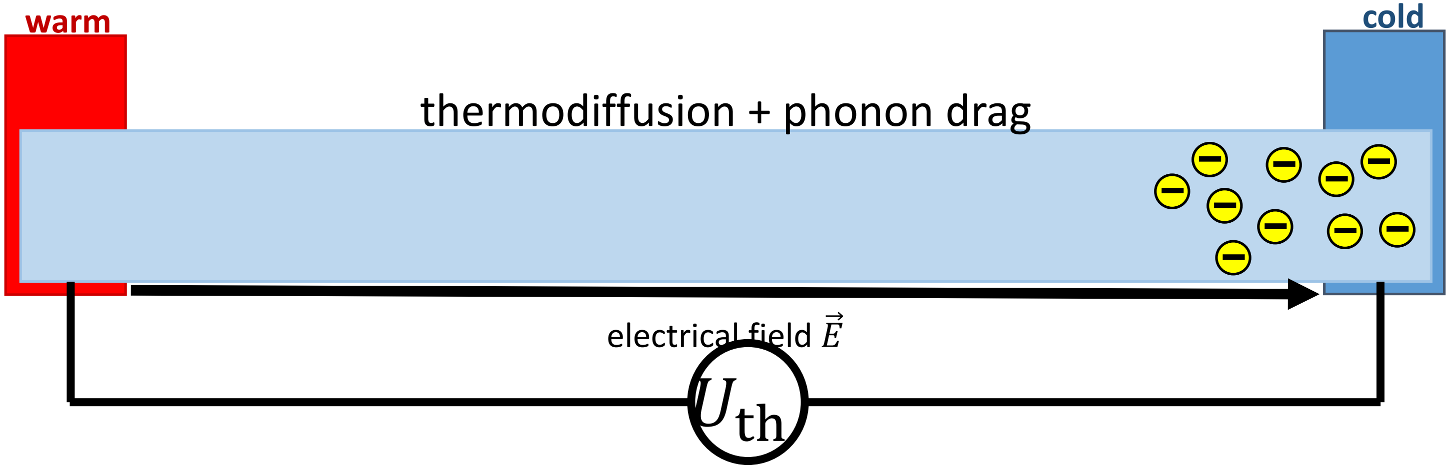
thermodiffusion: $$\boldsymbol{S_{\mathrm{d}} = -\frac{k_{\mathrm{B}}}{e}\left(r + \frac{5}{2} - \eta\right)}$$

Stratton, Phys. Rev. **126**, 2002 (1962).

phonon drag: $$\boldsymbol{S_{\mathrm{PD}} = -\frac{v^2}{T} \cdot \frac{1}{\mu_{\mathrm{AP}}} \cdot \tau_{\mathrm{Ph}}.}$$

Herring, Phy. Rev. **96**, 1163 (1954).

Hutson, JAP **32**, 2287 (1961).
Smith and Butcher, J. Physics: Cond. Mat. **2**, 2375–2382 (1990).



Phonon drag contribution to the Seebeck coefficient

phonon drag:

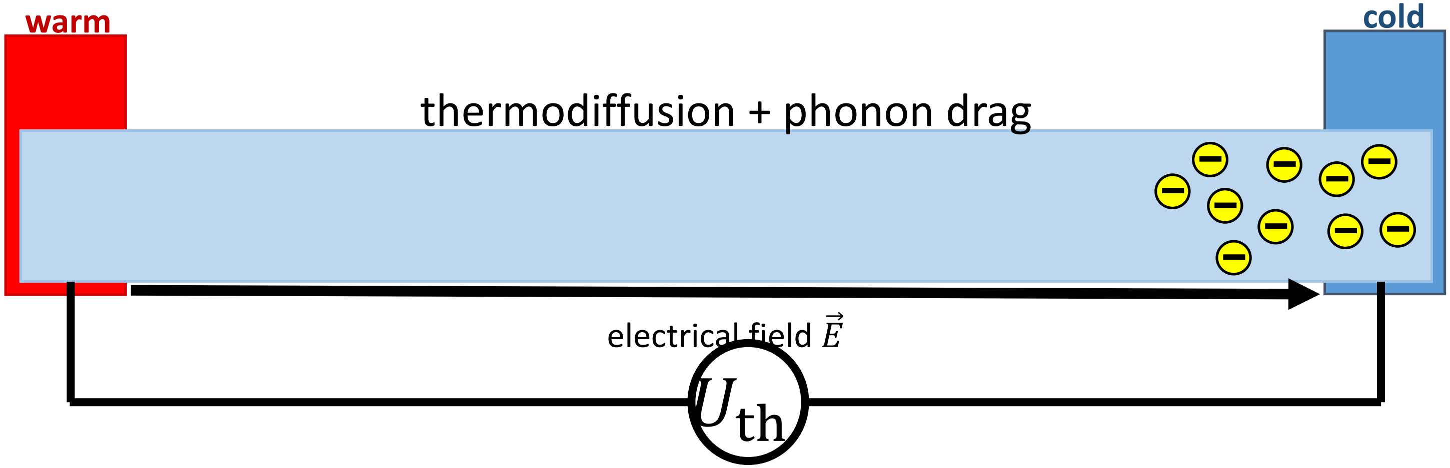
$$S_{\mathrm{PD}} = -\frac{v^2}{T} \cdot \frac{1}{\mu_{\mathrm{AP}}} \cdot \tau_{\mathrm{Ph.}}$$

$$= \frac{m^* v^2}{eT} \cdot \frac{\tau_{\mathrm{Ph.-Ph.}}}{\tau_{\mathrm{El.-Ph.}}}$$

Herring, Phy. Rev. **96**, 1163 (1954).

Hutson, JAP **32**, 2287 (1961).

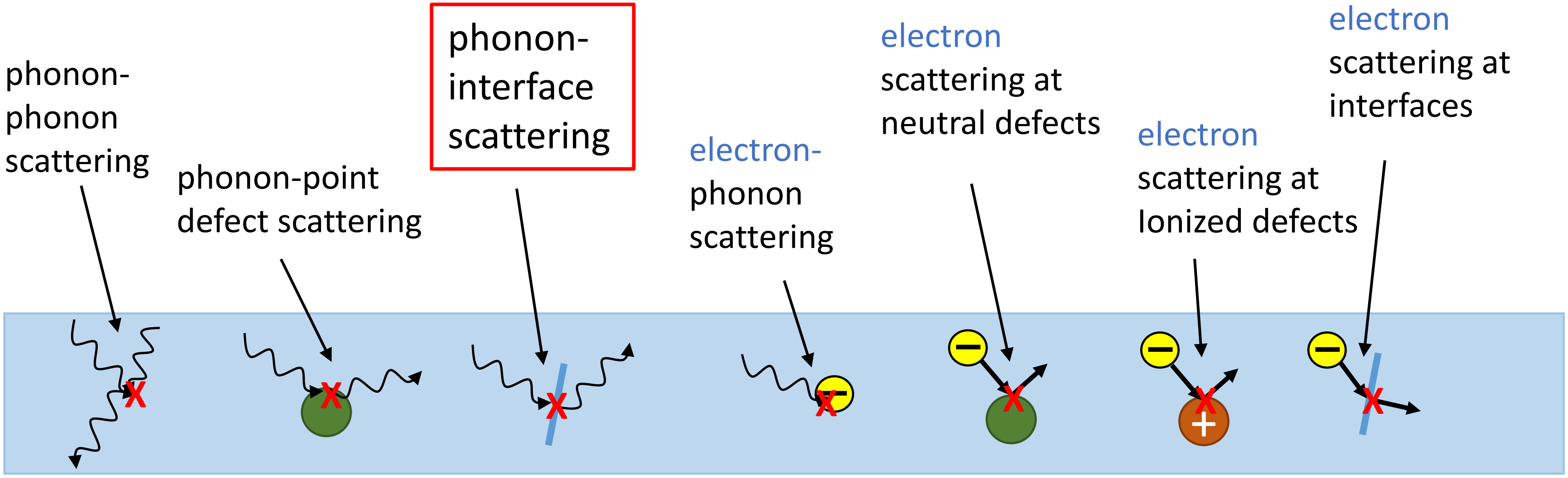
Smith and Butcher, J. Physics: Cond. Mat. **2**, 2375–2382 (1990).



Thermoelectricity: Full „zoo" of scattering mechanisms

phonon 〰→

electron ⊖



phonon-phonon scattering

phonon-point defect scattering

phonon-interface scattering

electron-phonon scattering

electron scattering at neutral defects

electron scattering at Ionized defects

electron scattering at interfaces

Reduced by…

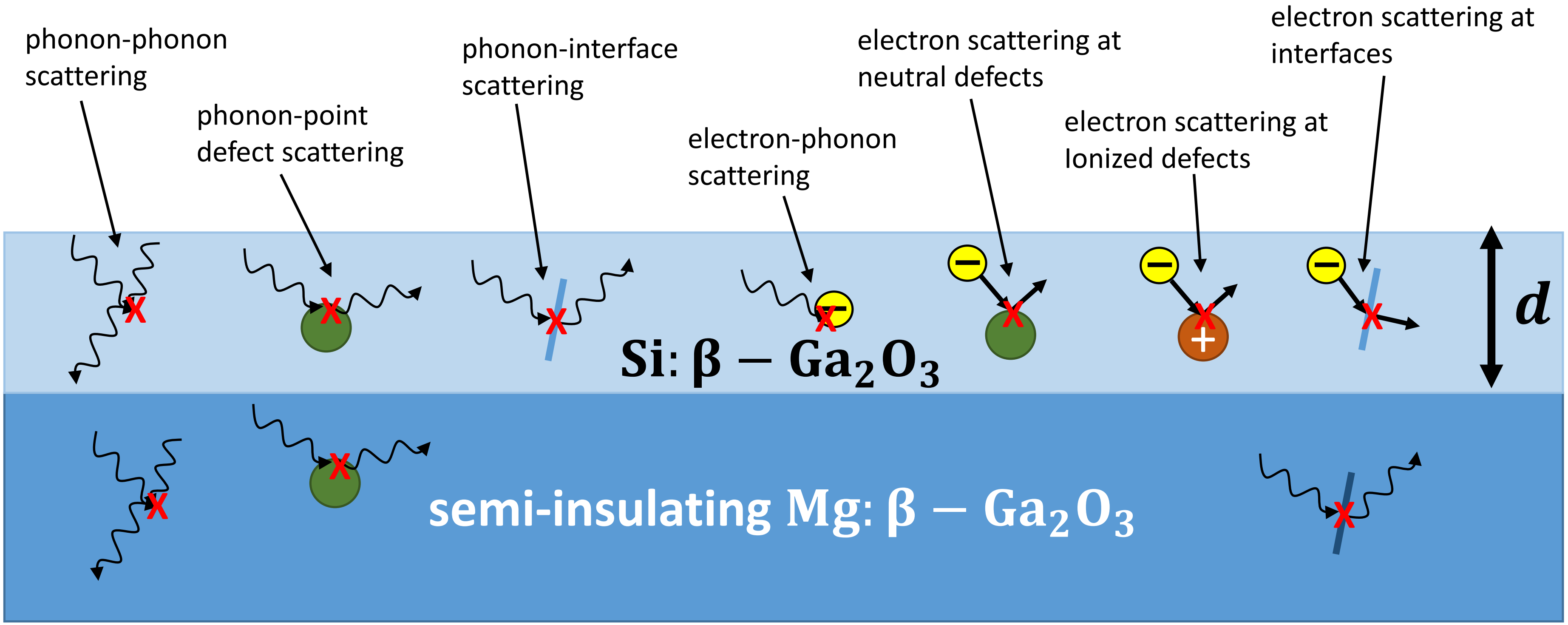
Temperature

Expitaxy

Homoepitaxy

Homoepitaxial films of ß-Ga2O3

→ Selection of in-plane phonons for phonon-drag effects by choosing a film thickness below the phonon mean-free path

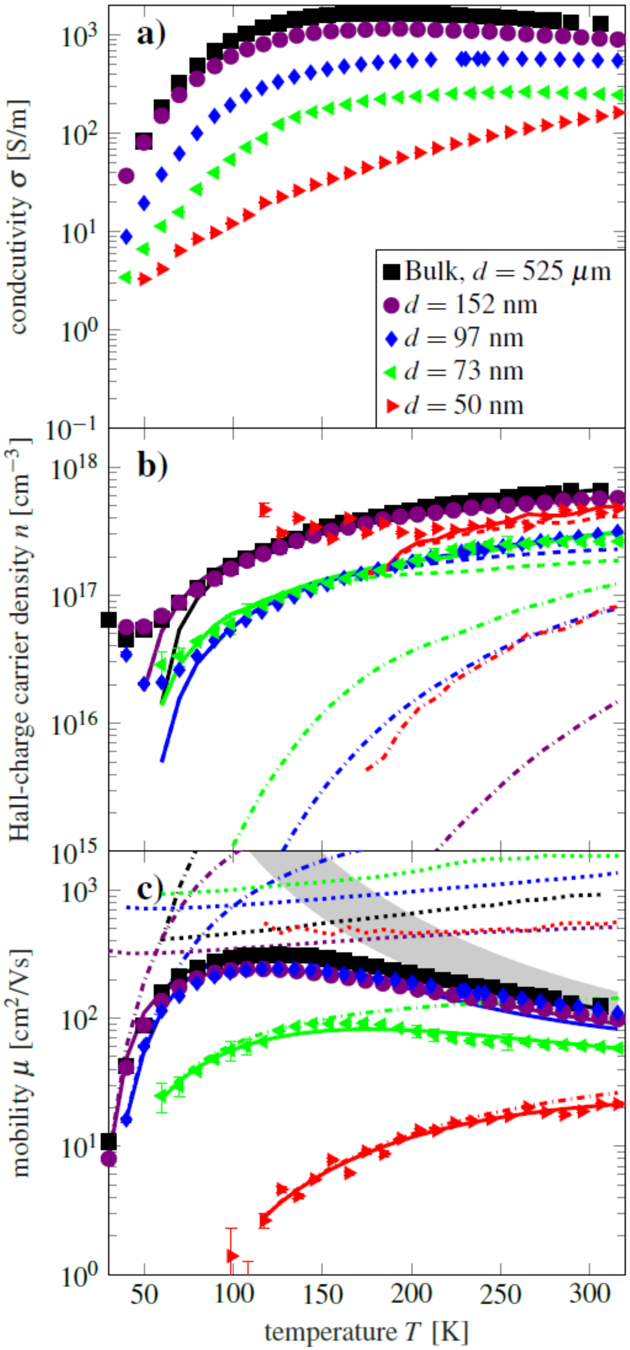


Homoepitaxial films of ß-Ga2O3:

Electrical conductivity

Charge carrier density

Mobility



J. Boy, *et al.*, *APL Mater.* **7**, 022526 (2019).

J. Boy, *PhD Thesis (2022)*

Homoepitaxial films of ß-Ga2O3:

reduced
chemical potential η

scattering factor *r*

thermodiffusion:

$$S_\mathrm{d} = -\frac{k_\mathrm{B}}{e}\left(r + \frac{5}{2} - \eta\right)$$

Stratton, Phys. Rev. **126**, 2002 (1962).

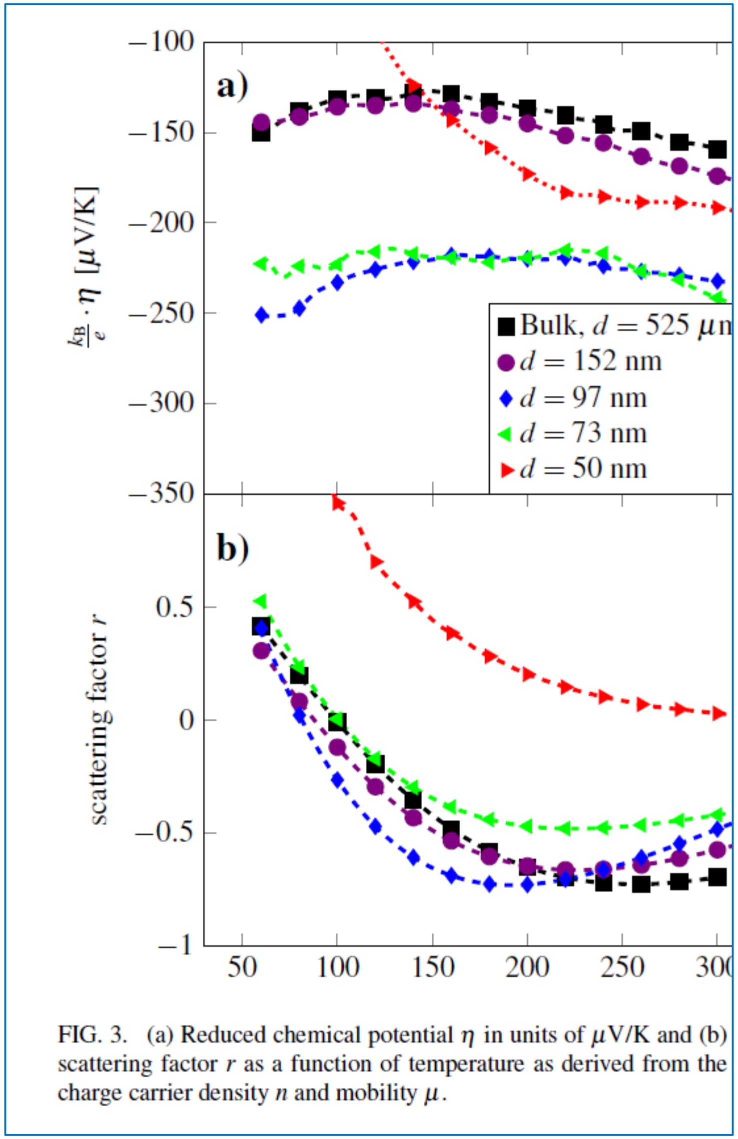
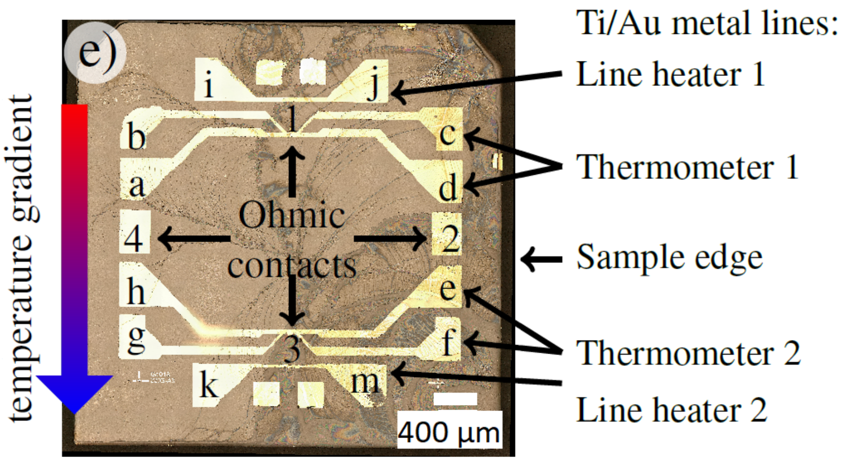


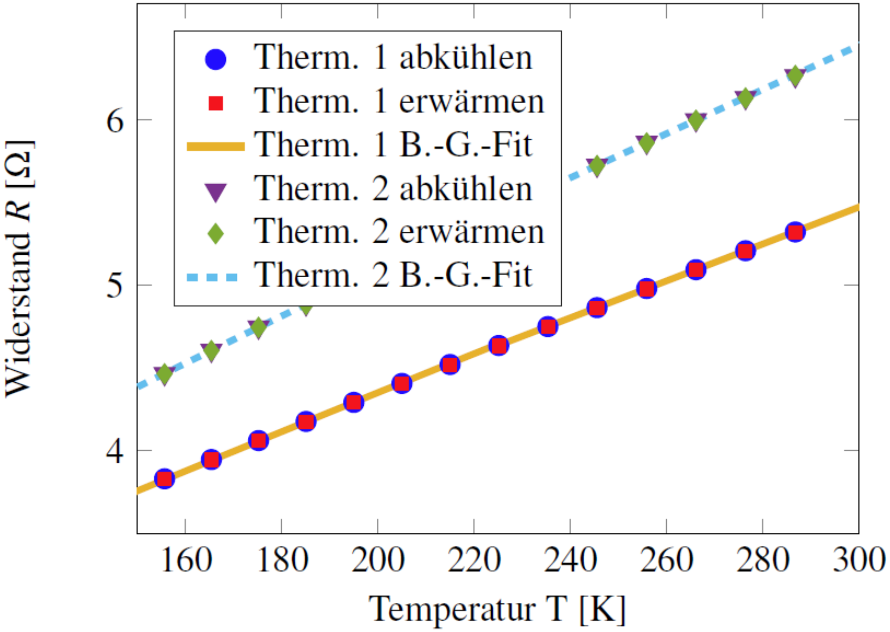
FIG. 3. (a) Reduced chemical potential η in units of μV/K and (b) scattering factor r as a function of temperature as derived from the charge carrier density n and mobility μ.

J. Boy, *et al.*, *APL Mater.* **7**, 022526 (2019).

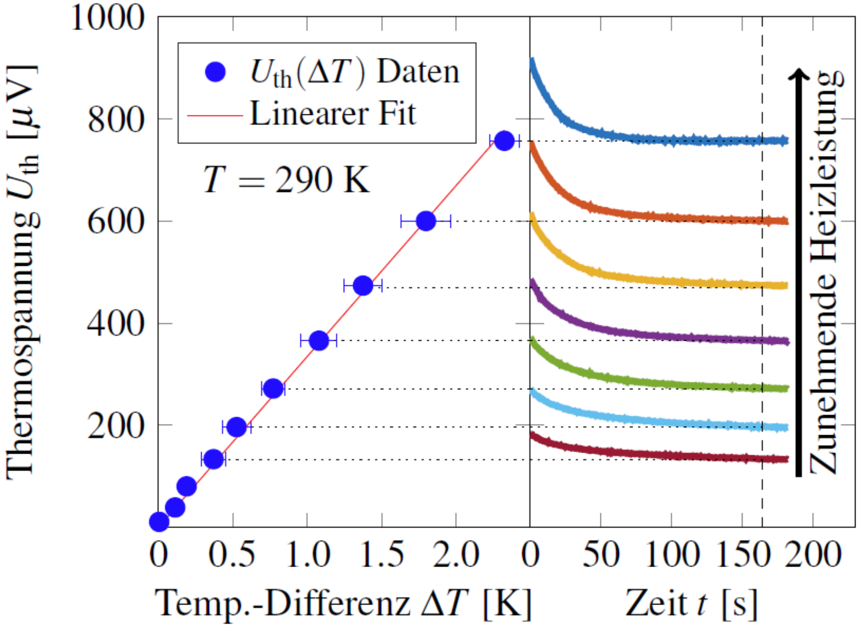
Thermoelectric micro measurement platform



Heater lines



Thermovoltages



J. Boy, *et al.*, *APL Mater.* **7**, 022526 (2019).

Giant phonon drag increase in $\beta - Ga_2O_3$ homoepitaxial thin films

$$S_{PD} = -\frac{v^2}{T}\frac{1}{\mu_{AP.}}\tau_{Ph.} = \frac{m^* v^2}{eT}\cdot\frac{\tau_{Ph.}}{\tau_{El.-Ph.}}$$

Calc. thermodiffusion part
fits well to theoretical work

Kumar and Singisetti, *APL* **117**, 262104 (2020).

[redacted]

Unpublished results.
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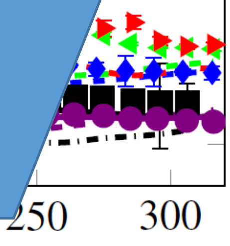

$$A = T \cdot S_{PD} \propto \frac{\tau_{Ph.}}{\tau_{El.-Ph.}}$$

thermodiffusion

J. Boy, *PhD Thesis (2022)*

Summary - Thermoelectric properties

Phonon-drag: A measure of electron-phonon interactions

- Thermoelectric voltages and Seebeck-coefficients

Phonon-transparent interfaces

- Thin film growth by homoepitaxy

Control of the effective electron-phonon interaction cross-section

- Film thickness below phonon mean-free path

➔ **Giant-phonon drag increase by design**

- selection of relevant in-plane phonons

Outlook: Results are generally valid for a wide range of materials.-

Transport properties of $\beta - Ga_2O_3$ single crystals and thin films

Anisotropic thermal conductivity & ballistic phonon transport

Handwerg, *et al.*, *Semicond. Sci. Technol.* 30, 024006 (2015).
Handwerg, *et al.*, *Semicond. Sci. Technol.* **31**, 125006 (2016).
R. Ahrling, *PhD Thesis (2023)*

Electrical properties & size effects of homoepitaxial thin films & flakes:

R. Mitdank, *et al.*, *Phys. Stat. Sol.*, A 211, 543-549 (2014).
R. Ahrling, *et al.*, *Scientific Reports* **9**, 13149 (2019).

Seebeck coefficients & (Giant-)phonon drag increase by thin film design

J. Boy, *et al.*, *APL Mater.* **7**, 022526 (2019).
J. Boy, *PhD Thesis (2022)*

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