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研究課題名（和文）マテリアルズ・インフォマティクスによる伝熱機能材料の設計

研究課題名（英文）Designing Thermal Functional Materials via Materials Informatics

研究代表者
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研究成果の概要（和文）：望ましい熱特性を備えた機能化材料を設計することは、熱交換器、熱界面材料、熱電、遮熱コーティング、および絶縁体の用途において重要です。このプロジェクトでは、ハイスループットスクリーニングやベイズ最適化などのマテリアルズインフォマティクス手法を開発して、マテリアルのシミュレーション/実験と機械学習ツールの間の熱機能通信を設計しました。目的関数、記述子の選択、目的関数を評価するための特性計算機、および情報学の最適化手法を設定することにより、高熱伝導率結晶、高選択性放射冷却材料、熱電膜、および磁気トンネル接合の探索に成功しました。この結果は、熱機能材料を設計するための材料情報学の利点をもたらしました。

研究成果の学術的意義や社会的意義
The target of this research is to develop high efficiency and novel materials informatics method for designing thermal functionalized materials. The developed method can be easily extended to other transport property designing, which is expected to contribute to both research and industry society.

研究成果の概要（英文）：Designing functionalized materials with desired thermal property holds its critical importance in applications of heat exchanger, thermal interface materials, thermoelectrics, thermal barrier coating and insulators. In this project, materials informatics methods including high-throughput screening and Bayesian optimization have been developed to design thermal functional materials. The key of designing thermal functional materials via materials informatics is to build communications between materials ' simulations/experiments and machine learning tools. By setting the objective function, descriptor selection, property calculator to evaluate the objective function, and an informatics optimization method, we have successfully explored the high thermal conductivity crystals, highly selective radiative cooling materials, thermoelectric films and magnetic tunnel junctions. Those results have shown great advantage of materials informatics to design thermal functional materials.

研究分野：熱工学

キーワード：マテリアルズ・インフォマティクス　伝熱機能材料　ベイズ最適化　機械学習

１．研究開始当初の背景

Designing functionalized materials with desired thermal property holds its critical importance in applications of heat exchanger, thermal interface materials, thermoelectrics, thermal barrier coating and insulators. However, two bottlenecks limit the designing efficiency: material selection and nanostructure designing. Selecting of most suitable material from tens of thousands of candidates is usually the first key question to face during the designing of thermal devices. The other one is more challenging and difficult: as the length scale of materials decreases to nanoscale, heat conduction becomes more controllable through manipulating the nanostructures. However, due to the various choice of structure parameters and coupled effect, it's rather difficult to obtain the optimal nanostructure quickly with desired thermal property from tremendous number of candidates. The key technology here is materials informatics (MI), which is a new interdisciplinary research to provide efficient tools to accelerate the materials discovery and design.

２．研究の目的

The target of this proposal is to develop high efficiency and novel materials informatics method, aiming to solve the two bottlenecks during the designing of thermal functionalized materials. The project not only realizes the goal of using informatics, but also aims to explore new mechanisms of heat transfer behind the designing or optimization result via informatics. The developed materials informatics method will not be limited to heat transport property, but can be easily extended to other transport property designing (such as electron, photon, and magnon transport), which is expected to contribute to both research and industry society.

３．研究の方法

The method mainly developed and adopted during this research project is materials informatics including high-throughput screening and Bayesian optimization. The key of designing thermal functional materials via materials informatics is to build communications between materials' simulations/experiments and machine learning tools [1]. By setting the objective function, descriptor selection, property calculator to evaluate the objective function, and an informatics optimization method, we have successfully explored the high thermal conductivity crystals, highly selective radiative cooling materials, thermoelectric films and magnetic tunnel junctions. Those results have shown great advantage of materials informatics to design thermal functional materials.

４．研究成果

(1) **Exploring high lattice thermal conductivity crystals via feature-based transfer learning**

Screening ultrahigh lattice thermal conductivity crystals via machine learning hold great importance since they play a critical role in the thermal management of electronic and optical devices. However, the lack of sufficient data to train a model is a serious hurdle. Transfer learning approach [2-3], as shown in Fig. 1, has been developed and employed to screen over 60000 compounds with phonon scattering phase space as the feature quantity and identified a set of semiconducting compounds with high thermal conductivities. The final, obtained materials in the top-14 list by feature-based and transfer learning screening all show high thermal conductivities, including boron arsenides (BAs), carbon (C), boron nitride (BN), and heterodiamond (BC_2N). They have thermal conductivities on the order of 1000 $Wm^{-1}K^{-1}$, validating the accuracy and high-efficiency of the developed screening method. Even though most materials in the top list are superhard materials, we reveal that superhard proper-ty do not necessarily lead to high lattice thermal conductivity. Large hardness means high elastic constants and group velocity of phonons in the linear dispersion regime, but the lattice

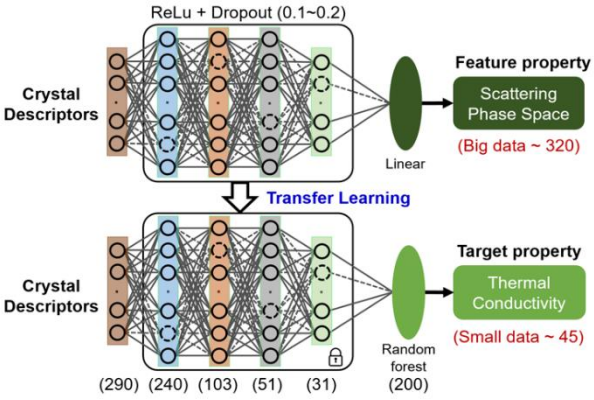

Fig. 1 Schematics of feature-based transfer learning.

thermal conductivity is determined also by other important factor such as the phonon relaxation time. What's more, the average or maximum dipole polarizability and the van der Waals radius are revealed to be the leading descriptors among those that can also be qualitatively related to anharmonicity.

Similar approach has also been used to explore the intrinsic origins of the hydrodynamic thermal transport and to find new materials interesting for science and engineering [4]. The hydrodynamic thermal transport is governed intrinsically by the hydrodynamic scale and the thermal conductivity. The correlations between these intrinsic properties and harmonic and anharmonic properties, and a large number of compositional (290) and structural (1224) descriptors of 131 crystal compound materials are obtained, revealing some of the key descriptors that determines the magnitude of the intrinsic hydrodynamic effects, most of them related with the phonon relaxation times. Then, a trained black-box model is applied to screen more than 5000 materials. The results identify materials with potential technological applications. Understanding the properties correlated to hydrodynamic thermal transport can help to find new thermoelectric materials and on the design of new materials to ease the heat dissipation in electronic devices. The key descriptors that determines the magnitude of the intrinsic hydrodynamic effects were revealed by machine learning, most of them related with the phonon relaxation times.

(2) **Design of a highly selective radiative cooling structure accelerated by materials informatics**

The thermal photonic structure for radiative cooling application has been designed by the method combining the rigorous coupled wave analysis and Bayesian optimization [5], as shown in Fig. 2. The structure with optimal thermal radiative property can be obtained by calculating only less than 1% of total candidate structures. The present work is the first trial using Bayesian optimization for radiative cooling device design and successfully tailored the thermal emittance selectively falling within the atmospheric window by hybrid grating and multilayer structures.

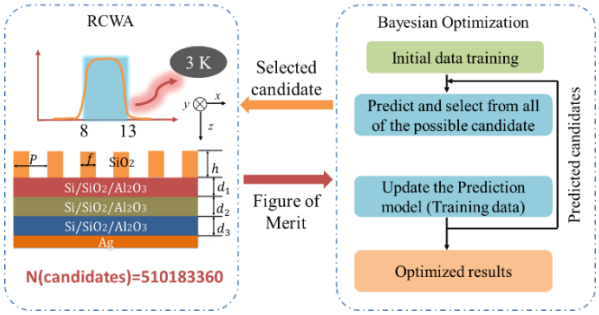

Fig. 2 Schematics of feature-based transfer learning.

(3) **Identifying optimal strain in bismuth telluride thermoelectric film by combinatorial gradient thermal annealing and machine learning**

To identify the optimal internal strain in Bi_2Te_3 that gives high thermoelectric performance, we developed a design framework that directly combines experimental measurements and machine learning, as illustrated in Fig. 3 [6]. XRD is widely used to analyze crystal structural information, including the lattice constants, grain size, and presence of defects. Meanwhile, thermoelectric characteristics, such as thermal conductivity and Seebeck coefficient, are readily collected through mapping. Herein, we focus on Seebeck coefficient because it shows the largest sensitivity on the strain as will be shown later. Using artificial neural network, we trained the relation between XRD and Seebeck coefficient based on collected pairs of XRD signals and Seebeck coefficient data. Based on the trained prediction model, we screen a large number of candidates of Bi_2Te_3 structures that are randomly generated to have different internal strain to identify the internal strain that gives large Seebeck coefficient.

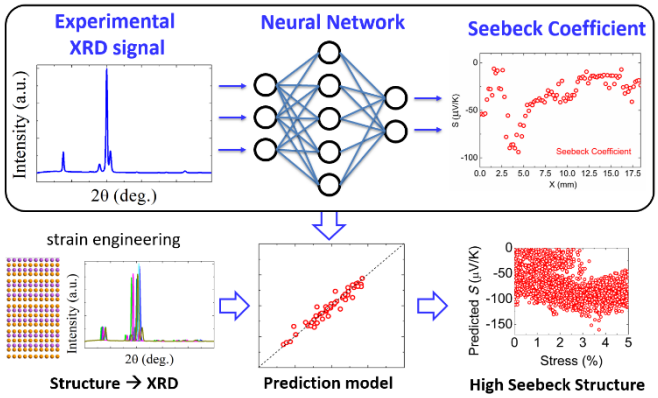

Fig. 3 Diagram of design thermoelectrics via machine learning.

(4) **Machine learning analysis of tunnel magnetoresistance of magnetic tunnel junctions with disordered $MgAl_2O_4$**

Through Bayesian optimization and the LASSO technique combined with first-principles calculations as shown in Fig. 4, we studied the tunnel magnetoresistance (TMR) of Fe/disordered-$MgAl_2O_4$(MAO)/Fe(001) magnetic tunnel junctions (MTJ) to determine structures of disordered-MAO that give large TMR ratios [7]. The optimal structure with the largest TMR ratio was obtained by Bayesian optimization with 1728 structural candidates, where the convergence was reached within 300 structure calculations. Characterization of the obtained structures suggested that the in-plane distance between two Al atoms plays an important role in determining the TMR ratio. Since the Al-Al distance of disordered MAO significantly affects the imaginary part of complex band structures, the majority-spin conductance of the Δ_1 state in Fe/disordered-MAO/Fe MTJs increases with increasing in-plane Al-Al distance, leading to larger TMR ratios. Furthermore, we found that the TMR ratio tended to be large when the ratio of the number of Al, Mg, and vacancies in the [001] plane was 2:1:1, indicating that the control of Al atomic positions is essential to enhancing the TMR ratio in MTJs with disordered MAO. The present work reveals the effectiveness and advantage of material informatics combined with first-principles transport calculations in designing high-performance spintronic devices based on MTJs.

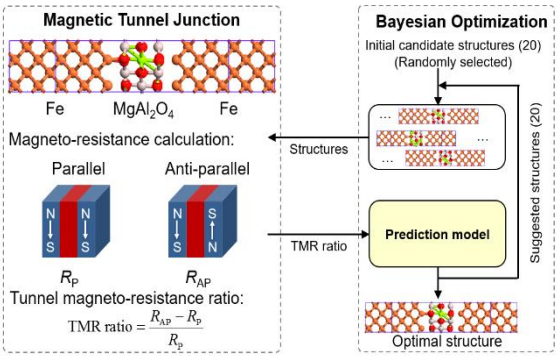

Fig. 4 Schematics of Bayesian optimization for magnetic tunnel junctions design.

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５．主な発表論文等

〔雑誌論文〕　計8件（うち査読付論文　8件／うち国際共著　8件／うちオープンアクセス　1件）

１．著者名 Sasaki Michiko、Ju Shenghong、Xu Yibin、Shiomi Junichiro、Goto Masahiro	４．巻 22
２．論文標題 Identifying Optimal Strain in Bismuth Telluride Thermoelectric Film by Combinatorial Gradient Thermal Annealing and Machine Learning	５．発行年 2020年
３．雑誌名 ACS Combinatorial Science	６．最初と最後の頁 782～790
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acscombsci.0c00112	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Ju Shenghong、Shimizu Shuntaro、Shiomi Junichiro	４．巻 128
２．論文標題 Designing thermal functional materials by coupling thermal transport calculations and machine learning	５．発行年 2020年
３．雑誌名 Journal of Applied Physics	６．最初と最後の頁 161102～161102
掲載論文のDOI（デジタルオブジェクト識別子） 10.1063/5.0017042	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Shao Cheng、Matsuda Kensuke、Ju Shenghong、Ikoma Yoshifumi、Kohno Masamichi、Shiomi Junichiro	４．巻 129
２．論文標題 Phonon transport in multiphase nanostructured silicon fabricated by high-pressure torsion	５．発行年 2021年
３．雑誌名 Journal of Applied Physics	６．最初と最後の頁 085101～085101
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１．著者名 Ju Shenghong、Miura Yoshio、Yamamoto Kaoru、Masuda Keisuke、Uchida Ken-ichi、Shiomi Junichiro	４．巻 2
２．論文標題 Machine learning analysis of tunnel magnetoresistance of magnetic tunnel junctions with disordered MgAl2O4	５．発行年 2020年
３．雑誌名 Physical Review Research	６．最初と最後の頁 23187
掲載論文のDOI（デジタルオブジェクト識別子） 10.1103/PhysRevResearch.2.023187	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

1．著者名 Guo Jiang、Ju Shenghong、Shiomi Junichiro	4．巻 45
2．論文標題 Design of a highly selective radiative cooling structure accelerated by materials informatics	5．発行年 2020年
3．雑誌名 Optics Letters	6．最初と最後の頁 343～343
掲載論文のDOI（デジタルオブジェクト識別子） 10.1364/OL.45.000343	査読の有無 有
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1．著者名 Yamada Hironao、Liu Chang、Wu Stephen、Koyama Yukinori、Ju Shenghong、Shiomi Junichiro、Morikawa Junko、Yoshida Ryo	4．巻 5
2．論文標題 Predicting Materials Properties with Little Data Using Shotgun Transfer Learning	5．発行年 2019年
3．雑誌名 ACS Central Science	6．最初と最後の頁 1717～1730
掲載論文のDOI（デジタルオブジェクト識別子） 10.1021/acscentsci.9b00804	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

1．著者名 Liu Jing、Ju Shenghong、Nishiyama Norimasa、Junichiro Shiomi	4．巻 100
2．論文標題 Anomalously low thermal conductivity in superhard cubic Si3N4	5．発行年 2019年
3．雑誌名 Physical Review B	6．最初と最後の頁 64303
掲載論文のDOI（デジタルオブジェクト識別子） 10.1103/PhysRevB.100.064303	査読の有無 有
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1．著者名 Feng Zhihao、Dong Huicong、Ju Shenghong、Wen Bin、Zhang Yuwen、Melnik Roderick	4．巻 21
2．論文標題 High bond difference parameter-induced low thermal transmission in carbon allotropes with sp2 and sp3 hybridization	5．発行年 2019年
3．雑誌名 Physical Chemistry Chemical Physics	6．最初と最後の頁 12611～12619
掲載論文のDOI（デジタルオブジェクト識別子） 10.1039/C9CP01029G	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計8件（うち招待講演　6件／うち国際学会　4件）

1．発表者名
Shenghong Ju, Ryo Yoshida, Chang Liu, Kenta Hongo, Terumasa Tadano, Junichiro Shiomi

2．発表標題
Exploring ultrahigh lattice thermal conductivity crystals via feature-based transfer learning

3．学会等名
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4．発表年
2020年

1．発表者名
Shenghong Ju

2．発表標題
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3．学会等名
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4．発表年
2020年

1．発表者名
Shenghong Ju

2．発表標題
Machine learning analysis of tunnel magnetoresistance of magnetic tunnel junctions with disordered MgAl2O4

3．学会等名
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4．発表年
2020年

1．発表者名
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2．発表標題
High Thermoelectric Performance in Metastable Phases of Silicon: A Density Functional Theory Study

3．学会等名
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4．発表年
2020年

１．発表者名
鞠 生宏，塩見 淳一郎

２．発表標題
マテリアルズ・インフォマティクスによる伝熱機能材料の設計

３．学会等名
第66回応用物理学会春季学術講演会

４．発表年
2019年

１．発表者名
Ju Shenghong

２．発表標題
Desing thermal functional materials via materials informatics

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The 4th Workshop on Thermal Transport, Wuhan, China, 2019（招待講演）

４．発表年
2019年

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Ju Shenghong

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４．発表年
2019年

１．発表者名
Ju Shenghong、Shiomi Junichiro

２．発表標題
Application of materials informatics on designing thermal functional materials

３．学会等名
The 6th Asian Materials Data Symposium, Shanghai, China, 2019（招待講演）（国際学会）

４．発表年
2019年

〔図書〕　計1件

1．著者名 Jiang Guo, Shenghong Ju and Junichiro Shiomi	4．発行年 2020年
2．出版社 IOP Publishing Ltd	5．総ページ数 488
3．書名 Chapter: Application of Bayesian optimization to thermal science, in Book titled Nanoscale Energy Transport: Emerging phenomena, methods and applications	

〔産業財産権〕

〔その他〕

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6．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

7．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計1件

国際研究集会 The 6th Asian Materials Data Symposium	開催年 2019年～2019年

8．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関