Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supplementary Information

Absence of phonon gap driven ultralow lattice thermal conductivity in half-Heusler LuNiBi

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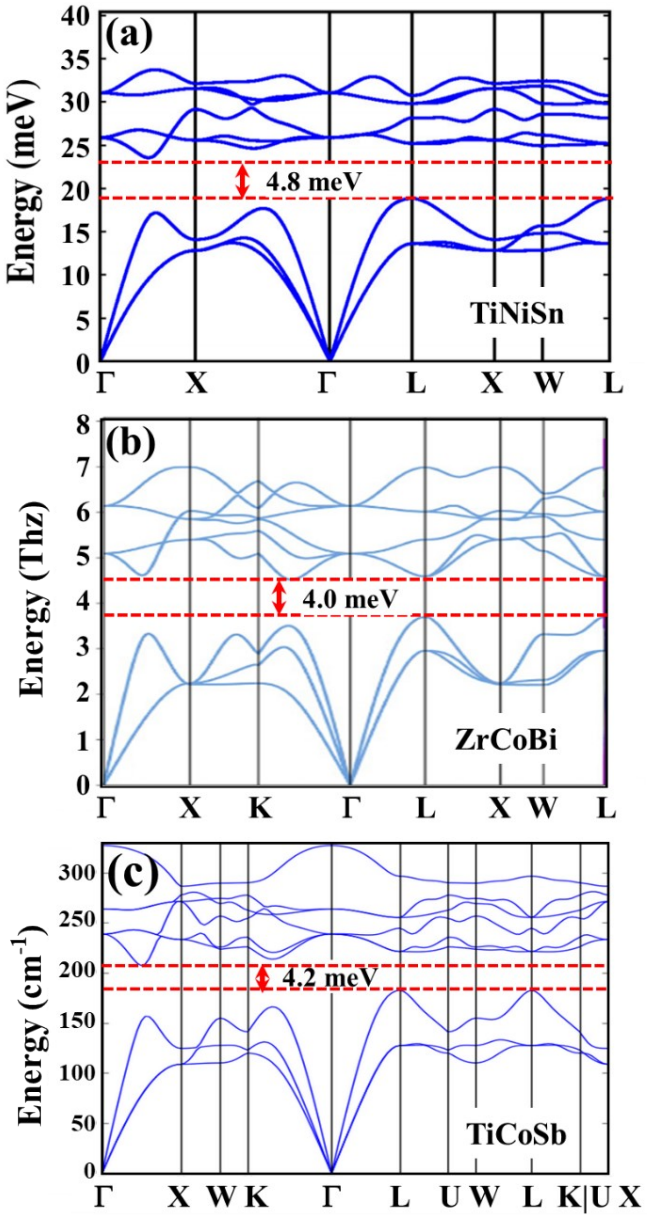

Figure S1. The phonon dispersion for half-Heusler of TiNiSn[1], ZrCoBi[2] and TiCoSb[3].

1. Two-channel Phonon Transport Model

To understand the physical mechanism of the two-channel phonon transport model, distribution of the mean free path (MFP) at room temperature for two half-Heusler compounds is shown in **Figure S2a**. The most prominent feature here is that a considerable portion of mean free path are smaller than the minimum atomic distance or the so-called Ioffe-Regel limit, which can be ascribed to the dual effect of short lifetime and small phonon velocity. This result suggests that the phonon Boltzmann transport equation methodology is no longer appropriate, since phonons are often considered to be "ill defined" in this case.[4, 5] According to two-channel phonon transport model, there is a coexistence of a phonon conduction channel (κ_{latt}) and the

hopping channel of localized vibrations (κ_{hop}) for the crystalline materials, particularly with loosely bound atoms. The former signifies the thermal transport carried by the "well-defined" phonon modes, whose MFP is larger than the lattice parameters, while the latter denotes the direct energy transfer between localized atomic vibrations, recognized as the "ill-defined" phonon modes, whose MFP is smaller than that of Ioffe-Regel limit. The "well-defined" phonon modes are commonly described by the Boltzmann transport equation methods. While the thermal conductivity of "ill-defined" phonon is calculated via the Cahill-Watson-Pohl model.[5] Note that the κ_{latt} obtained here does not subtract the contribution from phonons with MFPs less than the Ioffe-Regel limit (0.3 nm) both for LuNiBi and YNiBi because that the phonons with MFP less than the Ioffe-Regel limit give a negligible contribution for κ_{latt} as shown in **Figure S2b**.

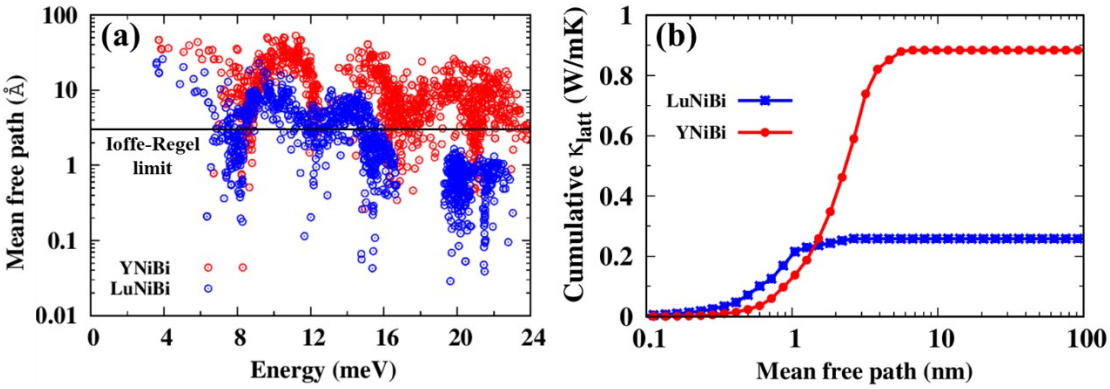

Figure S2. (a) Phonon frequency dependent mean free paths and (b) mean free paths dependent cumulative thermal conductivity for LuNiBi and YNiBi at room temperature. The black line marks the Ioffe-Regel limit defined by the average atomic distance in LuNiBi.

Table S1. Comparison of thermal conductivity at room temperature for Selected Materials in Fig.1a. The values are theoretical results except for explicit indication of "exp" in the table. (Unit: W/mK)

Structure	κ_{latt}	Structure	κ_{latt}
LuNiBi	0.26	Fe_2VAl	23.00[6], 28.00[7]exp
YNiBi	0.88	Ba_2AuBi	3.60[8]
HfRhSb	17.35[9]	Ba_2AuAs	4.70[8]
FeNbSb	18.00[10], 27.00[11]	Sr_2AuBi	4.20[8]
FeVSb	12.20[12]	Sr_2AuAs	5.40[8]
TiCoSb	25.00[13]	Ca_2AuBi	4.80[8]
TiNiSn	15.29[14], 7.80[15]exp	Ca_2HgPb	6.10[8]
ScNiSb	19.50[11]	$CoSb_3$	11.50[16]
ScNiBi	14.30[11]	$SrRu_4As_{12}$	0.47[17]exp
ZrNiSn	13.40[18], 5.90[19]exp	$CeFe_4As_{12}$	3.80[20]exp
ZrNiPb	13.87[14]	$BaCo_4Sb_{12}$	6.10[16]

ZrCoSb	23.06[21], 19.86[22]exp	$YbFe_4Sb_{12}$	0.34[23]
ZrCoBi	17.70[14], 9.00[24]exp	$K_8Al_8Si_{38}$	1.20[25]exp
LaPdSb	6.50[26]exp	Si_2Ge	0.28[27]
GdPdSb	4.90[26]exp	$Ba_8Ga_{16}Ge_{30}$	2.00[28]
HfNiSn	18.50[14]	BiCuSeO	0.55[29]
TaIrSn	18.80[30]	$NaCo_2O_4$	3.94[31]
TaFeSb	28.15[14]	$SrHfO_3$	4.54[32]exp
TaCoSn	19.75[14]	$SrRuO_3$	5.72[32]exp
TiPdS	5.47[33]	PbTe	1.50[34], 2.00[35]exp
NbCoSn	6.50[36]exp	SnSe	0.72(*b*)[37]exp

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