Supporting Information

**Prediction of higher thermoelectric performance in BiOCuSe by weakening electron-polar optical phonon scattering**

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**Supplementary Figures**



**Supplementary Figure 1 | Lattice thermal conductivity of BiOCuSe as reproduced from literature,1 which is used to calculate thermoelectric figure of merit ZT of BiOCuSe in this work.**



**Supplementary Figure 2 | The variation of thermoelectric charge transport properties with carrier concentration. The calculated results as compared to that by excluding PF with frequency > 200 cm-1 of (a) Seebeck coefficient, (b) electrical conductivity, (c) electrical thermal conductivity. The calculated results as compared to that by excluding PF with frequency < 200 cm-1 of (d) Seebeck coefficient, (e) electrical conductivity, (f) electrical thermal conductivity.**



**Supplementary Figure 3 | The variation of (a) power factor and (b) ZT of BiOCuSe as a function of carrier concentration and temperature after excluding the scattering effects of PO phonon with frequency > 200 cm-1. The variation of (c) power factor and (d) ZT of BiOCuSe as a function of carrier concentration and temperature after excluding the scattering effects of PO phonon with frequency < 200 cm-1.**

**Supplementary Tables**

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| **Supplementary Table 1 | Longitudinal optic phonon frequencies of BiOCuSe in cm-1** |
| LO Modes | This work | Literature2 | Experiment3 |
| Eu | 56.2 | 61.2 |  |
| A2u | 91.4 | 89.4 | 92.0 |
| Eu | 135.9 | 142.7 |  |
| A2u | 175.2 | 178.6 |  |
| Eu | 341.9 | 347.7 |  |
| A2u | 472.9 | 473.7 | 475.0 |

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| **Supplementary Table 2 | Born effective charge of BiOCuSe** |
|  | This work | Literature2  |
| $$Z\_{Bi}^{\*}$$ | In plane | 6.50 | 6.46 |
| Out of plane | 5.94 | 5.93 |
| $$Z\_{O}^{\*}$$ | In plane | -4.28 | -4.28 |
| Out of plane | -4.36 | -4.42 |
| $$Z\_{Cu}^{\*}$$ | In plane | 1.46 | 1.44 |
| Out of plane | 1.09 | 1.06 |
| $$Z\_{Se}^{\*}$$ | In plane | -3.67 | -3.67 |
| Out of plane | -2.67 | -2.70 |
| $$k\_{\infty }$$ | In plane | 17.61 | 18.01 |
| Out of plane | 13.25 | 13.79 |

**Supplementary References**

1. Ren, G. K. *et al.* Enhanced thermoelectric properties in Pb-doped BiCuSeO oxyselenides prepared by ultrafast synthesis. *RSC Adv.* **5**, 69878–69885 (2015).

2. Saha, S. K. Exploring the origin of ultralow thermal conductivity in layered BiOCuSe. *Phys. Rev. B* **92**, 041202 (2015).

3. Berdonosov, P. S. Powder X-Ray and IR Studies of the New Oxyselenides *M*OCuSe (*M* = Bi, Gd, Dy). *J. Solid State Chem.* **118**, 74 (1995).