Discovering 2D Semiconductors with High Intrinsic Carrier Mobility at Room Temperature

Chenmu Zhang, Ruoyu Wang, Himani Mishra, Yuanyue Liu*

Texas Materials Institute and Department of Mechanical Engineering,
The University of Texas at Austin, Austin, Texas 78712, USA
Yuanyue.liu@austin.utexas.edu

Abstract:
Two-dimensional (2D) semiconductors have demonstrated great potential for next-generation electronics and optoelectronics. However, the current 2D semiconductors suffer from intrinsically low carrier mobility at room temperature, which significantly limits its applications. Here we discover a variety of new 2D semiconductors with mobility one order of magnitude higher than the current ones. The discovery is made by developing effective descriptors for high-throughput computationally screening of the 2D materials database, followed by using state-of-the-art first principles method to accurately calculate the mobility. Further analyses attribute their exceptional mobilities to small effective mass, high sound velocity, high optical phonon frequency, small ratio of Born charge vs. polarizability, and/or weak electron-phonon coupling. Our work opens up new materials to realize high device performance and/or exotic physics that are difficult to achieve previously, and improves the understanding of the carrier transport mechanism.

Main text:
One of the grand challenges for electronic materials research is to find an alternative to Si with a suitable band gap, high carrier mobility at room temperature, and ambient stability, when thinning down to atomic thickness (for efficient gate control). The current candidates all suffer from one or more problems. For example, although graphene has very high carrier mobility, it does not have band gap. 2D crystalline semiconductors, on the other hand, despite their many useful properties (e.g. efficient gate control [1,2], optical transparency [3], and mechanical flexibility [4,5]), currently suffer from intrinsically low carrier mobility at room temperature, due to strong scattering by phonons. For example, MoS$_2$, one of the most common 2D semiconductors, has an intrinsic electron mobility $[6,7] < 200$ cm$^2$V$^{-1}$s$^{-1}$, much lower than electron mobility of bulk silicon ($1400$ cm$^2$V$^{-1}$s$^{-1}$). Tremendous efforts have been devoted to search for higher-mobility 2D semiconductors. The past few years have witnessed the rise of 2D black phosphorus [8], indium selenide [9], etc., each of which has attracted great interest. However, their mobilities are still not satisfactory.

There exist thousands of 2D materials which are not/less studied. To evaluate their potential as high-mobility semiconductors, here we explore the Computational 2D Materials Database (C2DB) [10,11]. We first formulated a set of descriptors based on the effective mass, Fröhlich scattering, and acoustic deformation potential scattering, and used them to narrow down the candidates. Then we apply state-of-the-art first-principles method to accurately calculate the intrinsic mobility. We discover a number of semiconductors with extremely high mobility, for example: BSb (electron
mobility: ~5000; hole: ~ 7000 cm²V⁻¹s⁻¹), ZrI₂ (hole: ~ 5000 cm²V⁻¹s⁻¹), and Ga₂Ge₂Te₂ (electron: ~ 2000 cm²V⁻¹s⁻¹; its 3D counterpart exists experimentally in a van der Waals layered form [12,13]). To understand the origins of their high mobilities, we analyze the effects of electronic structure, density of scatterings [14], and electron-phonon coupling strength for different phonon modes. We find that the high mobility can arise from small effective mass, high sound velocity, high optical phonon frequency, small ratio of Born charge vs. polarizability, and/or weak electron-phonon coupling. The discovered materials as well as the mechanistic insights bring us a step closer to the next-generation electronics/optoelectronics.

Although the intrinsic mobility can be accurately calculated based on the Boltzmann transport theory in conjunction with density functional perturbation theory (DFPT) [15–21], applying it to every candidate in the database is too computationally expensive. To make the discovery more efficient, we first use several “descriptors” that can be easily calculated, to narrow down the candidates.

The first descriptor is the “combined effective mass” $M$, which we define as:

$$M = \sqrt{m^*_i m^*_d}$$

(1)

where $m^*_i$ is the effective mass along carrier transport direction, and $m^*_d$ is the density of state effective mass that can be approximated by $N \sqrt{m^*_i m^*_j}$ (here $N$ is the degeneracy of conduction/valence band extremes, and $x$ and $y$ are the two directions defined by the database). We use this descriptor because:

1. Previous studies have shown that a low density of electronic states can suppress the scattering, as there are less states available for carriers to be scattered to. For example, the materials with CBM/VBM located at Gamma point [22], or having two spin-polarized CBMs/VBMs at K and K' points of the hexagonal lattice [23], could host high mobility due to the absence of inter-valley scattering. Thus we include the density of states information through $m^*_d$ in the $M$.

2. According to the Drude model, when the scattering is fixed, decreasing the effective mass along the transport direction can improve the mobility. Therefore, an effective mass specific to the transport direction $m^*_i$ has been included in the $M$. In order to evaluate the maximum mobility for anisotropic material, we use the smallest $m^*_i$ available in the C2DB database for a given material.

The $M$ is a qualitative descriptor for mobility. To quantitatively estimate the mobility, we consider two important scatterings: Fröhlich scattering and acoustic deformation potential (ADP) scattering. The Fröhlich scattering originates from the interaction between carriers and the long-range dipolar potentials generated by optical phonons. It is the dominant scattering in many polar semiconductors [19,24,25]. We assume its $g$ has the form:

$$g_j^\kappa(q) = \frac{i e^2}{2\Omega} \sum_k \left( \frac{\hbar}{2 M_k \omega_j} \right)^{1/2} \frac{q \cdot Z_{\kappa} \cdot e_{\kappa,j}(q)}{1 + 2 \pi \alpha_{\kappa} \omega_j |q|}$$

(2)

where $q$ is the phonon wavevector and $q$ is its unit vector, $\Omega$ is the area of unit cell, $\kappa$ is the index of atom in the unit cell, $M_k$ is the atomic mass; $j$ is the index of the optical phonon mode, and $\omega_j$ is its phonon frequency (approximated by a special constant obtained using the approach of Ref. [26]), $e$ is
the phonon eigenvector; $Z^*$ is the Born effective charge, and $\alpha_{2D}$ is the in-plane polarizability of the 2D crystal (approximated by $\alpha_{2D} = (\alpha_{2D,xx} + \alpha_{2D,yy})/2$). See Note S4 in the SM for calculation details. With the $g$ in hand, we can obtain the scattering rates via Eq. S3, and further the mobility due to the Fröhlich scattering ($\mu_F$) via Eq. S1. This is done numerically, under the assumptions of a parabolic electronic band (with effective mass $m^* = (m^*_{xx} + m^*_{yy})/2$) and multiple dispersion-less phonon modes (with frequency $\omega_0$).

The ADP scattering originates from the coupling of the electrons with LA phonons. The corresponding mobility can be estimated by [27]:

$$\mu_{ADP} = \frac{e\hbar^3 \rho v_{LA}^2}{k_B T m^* \alpha_{2D} (D_{LA})^2} = \frac{e\hbar^3 C_{2D}}{k_B T m^* \alpha_{2D} (D_{LA})^2},$$

(3)

where $\rho$ is the area density, $v_{LA}$ is the longitudinal sound velocity, and $C_{2D}$ is the 2D elastic modulus (approximated by $C_{2D} = (C_{2D,xx} + C_{2D,yy})/2$).

![Figure 1](image-url)

**Figure 1** (a) Illustration of screening procedures to discover 2D semiconductors with potential high carrier mobility. (b) Crystal structures for representative 2D semiconductors with mobilities over 1000 cm²V⁻¹s⁻¹.

The mobility with both Fröhlich and ADP scatterings can be obtained from the individual mobilities following the Matthiessen’s rule:

$$\frac{1}{\mu_{up}} = \frac{1}{\mu_{F}} + \frac{1}{\mu_{ADP}}$$

(4)
Since only two types of scatterings are considered in Eq. 4, the $\mu_{up}$ should be regarded as the mobility upper limit, and can be used to exclude the low-mobility materials. The physical parameters needed for $M$, $\mu_{ADP}$, $\mu_{F}$ and $\mu_{up}$ can be quickly extracted from the C2DB database, making them suitable for high-throughput screening. Note that there are other scatterings (e.g. piezoelectric scattering, optical deformation potential scattering). However, their $g$ are relatively difficult to obtain, which makes it difficult to formulate the corresponding descriptors that can be easily calculated.

Using these descriptors together with the band gap and stability (as shown in Fig. 1a and detailed in Note S3), we identified 81 2D semiconductors and accurately calculated their intrinsic mobilities. Fig. 2 shows their mobilities vs band gaps. There exist many 2D semiconductors with electron/hole mobility higher than that of MoS$_2$ electron. Particularly, we find the following materials with mobility > 1000 cm$^2$/V·s, all having a hexagonal lattice (and thus isotropic mobilities) and can be grouped into 5 types based on the atomic structure (as shown in Fig. 1b): (1) BSb (e: 5167; h: 6935h; ‘e’ for electron and ‘h’ for hole; in the unit of cm$^2$/V·s), BAs (e: 1524; h: 2439), InN (e: 2106), BP (e: 1151; h: 1921), InAs (e: 1001). The BSb, BAs, InN and BP are composed of III-V elements, and are atomically flat (similar to h-BN). (2) ZrI$_2$ (h: 5138), HfI$_2$ (h: 4782). The ZrI$_2$ and HfI$_2$ have a similar crystal structure to 2H phase MoS$_2$ with the metal layer in the middle. (3) Sn$_2$H$_2$ (e: 3227; h: 2063), Ge$_2$H$_2$ (e: 2791). The Sn$_2$H$_2$ and Ge$_2$H$_2$ are H functionalized IV elements in sp$^3$ configuration. (4) Sb$_2$ (h: 2044), As$_2$ (h: 1216). The Sb$_2$ and As$_2$ have a buckled hexagonal structure. (5) Ga$_2$Ge$_2$Te$_2$ (e: 1996). The Ga$_2$Ge$_2$Te$_2$ has a unique sextuple layered structure with atomic layers in order of Te-Ga-Ge-Ge-Ga-Te.

To extend the search, we also substitute the elements in the above-mentioned materials by those in the same group (see Note S7 in SM for details). We find several additional high mobility 2D semiconductors outside the database: AlBi (e: 2835; h: 3446) and GaSb (e: 1809; h: 1080) with crystal structures like BSb, and Al$_2$Ge$_2$Te$_2$ (e: 2023) whose structure is similar to Ga$_2$Ge$_2$Te$_2$. Their mobilities and HSE band gaps are added into Fig. 2.

We note that the mobilities of BSb (e and h), ZrI$_2$ (h), HfI$_2$ (h), AlBi (e and h), Sn$_2$H$_2$ (e and h), Ge$_2$H$_2$ (e), BAs (e and h), Sb$_2$ (h), Ga$_2$Ge$_2$Te$_2$ (e), Al$_2$Ge$_2$Te$_2$ (e), BP (h), InN (e) and GaSb (e) (13 materials in total) are higher than that of Si electron. Particularly, BSb, AlBi, Sn$_2$H$_2$ and BAs have both high electron mobility and hole mobility, as characterized by the ambipolar mobility $\mu_a$ (defined by $\mu_a = 2\mu_e\mu_h/($$\mu_e$+$\mu_h$) where $\mu_e$ and $\mu_h$ are electron and hole mobility respectively): 5922 for BSb, 3111 for AlBi, 2517 for Sn$_2$H$_2$ and 1876 for BAs. These excellent properties make them especially promising for electronic devices.
Figure 2 Mobility vs. band gap (HSE) for various 2D semiconductors. For comparison, the electron mobilities for MoS$_2$ and bulk Si are marked.

In order to understand why the mobilities are so high for those materials, we calculate the “Drude effective mass” ($\bar{m}^*$) and “Drude scattering rate” ($1/\bar{\tau}$) (see Note S9 in SM for definitions) [22]. The $\bar{m}^*$ is fully determined by the electronic structure and its occupation, while the information about phonons and the EPCs are wrapped in $1/\bar{\tau}$. Fig. 3a shows the $1/\bar{\tau}$ vs. $\bar{m}^*$ for the materials with e/h mobility > 1000 cm$^2$V$^{-1}$s$^{-1}$, as well as those for MoS$_2$ for comparison. We find that they all have $\bar{m}^*$ smaller than MoS$_2$, and $1/\bar{\tau}$ lower than MoS$_2$, which together give rise to their high mobilities. However, the contributions from $1/\bar{\tau}$ and $\bar{m}^*$ are different. For example, the $1/\bar{\tau}$ of Ga$_2$Ge$_2$Te$_2$ electron is only 50% of MoS$_2$ (15 vs. 30 ps$^{-1}$), while its $\bar{m}^*$ is 13.6% of MoS$_2$ (0.058 vs. 0.43 $m_e$), which is the major contributor to its ~15 times higher mobility than MoS$_2$ (1996 vs. 136 cm$^2$V$^{-1}$s$^{-1}$). In contrast, ZrI$_2$ has a comparable $\bar{m}^*$ (0.40 $m_e$) to that of MoS$_2$ (0.43 $m_e$), but its $1/\bar{\tau}$ is much lower (0.85 vs. 30 ps$^{-1}$), resulting in 38 times higher mobility than MoS$_2$ (5138 vs. 136 cm$^2$V$^{-1}$s$^{-1}$). BSb has both a small $\bar{m}^*$ (0.09 $m_e$ for hole and electron) and a low $1/\bar{\tau}$ (2.8 ps$^{-1}$ for hole and 3.6 ps$^{-1}$ for electron), which together make it the highest mobility material.
Figure 3 (a) Drude scattering rate and Drude effective mass for high-mobility (> 1000 cm²V⁻¹s⁻¹) 2D semiconductors. The lines are the iso-mobility contours. (b) Scattering rates of three representative high-mobility materials. For comparison, MoS₂ data are also shown.

To further understand why scattering rate can be so low in some materials, we focus on 3 representative materials: BSb, ZrI₂, and Ga₄Ge₂Te₂. We choose them because: (1) BSb and ZrI₂ are the first two highest mobility 2D semiconductors; (2) There exists vdW layered bulk Ga₄Ge₂Te₂, suggesting that its 2D form may be easy to experimentally realize [12,13]. Fig. 3b compares the state-dependent scattering rates for these 3 materials and MoS₂, which shows the order: MoS₂ > Ga₄Ge₂Te₂ ≫ BSb > ZrI₂, consistent with that seen from 1/τ.

To gain further insights, we decompose the scattering rate into contributions from individual phonon modes. We focus on the LA, TA, and LO modes, as they are often the dominant scattering sources to the intrinsic mobility. The top panels of Fig. 4 compare the mode-resolved scattering rates for different materials. Interestingly, compared with MoS₂, Ga₄Ge₂Te₂ has a stronger LO scattering, while weaker LA and TA scatterings. While for BSb and ZrI₂, the scatterings are weaker for all phonon modes.

To further understand the variation in the mode-resolved scattering, we calculate the mode-resolved “density of scatterings” (Dₛ; see Note S10 in SM for definition) [14] and the matrix elements for representative EPCs. The Dₛ is determined by the “match” between electron and phonon structures as well as their occupations, without involving the EPCs. It characterizes the density of scattering events for an initial state nk by a specific phonon mode v. A larger optical phonon frequency or a higher sound velocity would reduce the phonon populations, and thus lower the Dₛ, thereby resulting in a lower scattering rate and hence a higher carrier mobility. Similarly, a smaller effective mass would reduce the density of electronic states, which leads to a lower Dₛ. For the representative EPCs, we consider the |g|² between the initial state located at 30 meV above/below the CBM/VBM along Γ-M direction, and the final states at the iso-energy circle of the same valley.
Figure 4 Phonon-mode-resolved scattering rates (top panels), density of scatterings (middle panels), and representative (squared) EPC strengths (bottom panels) for BSb hole, ZrI$_2$ hole, and Ga$_2$Ge$_2$Te$_2$ electron, in comparison with MoS$_2$ electron. See the text for the definition of density of scatterings and the choice of the electronics sates for representative EPCs.

Fig. 4 shows the $D^S$ and the representative $|g|^2$ for LO, TA and LA modes across different materials. Although Ga$_2$Ge$_2$Te$_2$ has a lower $D^S$ than MoS$_2$ for the LO mode due to its smaller effective mass (0.05 vs. 0.5 $m_e$), it has larger $|g|^2$, leading to the stronger LO scattering. The larger LO $|g|^2$ in Ga$_2$Ge$_2$Te$_2$ can be explained by its larger ratio of the (maximum) Born charge to the in-plane polarizability: $R_{B/P}$=0.23 ($R_{B/P}$=0.07 in MoS$_2$). For LA and TA modes, the $D^S$ of Ga$_2$Ge$_2$Te$_2$ is comparable to that of MoS$_2$, while the $|g|^2$ is smaller, resulting in the weaker LA and TA scatterings. In contrast to Ga$_2$Ge$_2$Te$_2$, ZrI$_2$ has a larger $D^S$ than MoS$_2$ for LO mode resulting from its smaller LO frequency (15 vs. 47 meV in MoS$_2$), however, its $|g|^2$ is smaller thanks to the vanishing $R_{B/P}$ ($1.7 \times 10^{-4}$, which is mainly caused by the small maximum Born charge: 0.0012 vs. 0.5 in MoS$_2$), causing its nearly vanishing LO scattering. The $|g|^2$ of LA and TA modes in ZrI$_2$ are also smaller than those in MoS$_2$, giving rise to its smaller LA and TA scatterings. For BSb, although it has the largest LO $|g|^2$ because of its largest $R_{B/P}$ (0.51) among the 4 materials in comparison here, its LO $D^S$ is extremely small due to the highest LO phonon frequency (88 meV), therefore it has a weak LO scattering. BSb also has lower $D^S$ for LA and TA modes than
MoS$_2$ owing to its relatively large transverse/longitudinal sound velocity, which together with the smaller $|g|^2$ lead to its weaker LA and TA scatterings.

The above analyses suggest several basic and easy-to-calculate physical properties that are important to the mobility: small effective mass, high sound velocity, high optical phonon frequency, and small ratio of Born charge vs. polarizability. Indeed, as shown in Fig. S3, all the high-mobility materials (>1000 cm$^2$/V·s) possess one or a few of these properties, highlighting their effects on carrier transport.

In summary, we discovered a number of high-mobility 2D semiconductors and explained the origins of their high mobilities. We used the criteria of bandgap, stability, Fröhlich and acoustic deformation potential scatterings to screen the C2DB database, and then accurately calculated the mobility for 81 selected materials. We found 11(10) 2D semiconductors with electron (hole) mobility >1000 cm$^2$/V·s. Particularly, BSb has both high electron (5167 cm$^2$/V·s) mobility and high hole mobility (6935 cm$^2$/V·s). ZrI$_2$ has a high hole mobility (5138 cm$^2$/V·s) with bulk counterpart experimentally existing in vdW (or weakly interacting) layered form. Exemplified by BSb, ZrI$_2$ and Ga$_2$Ge$_2$Te$_2$, we uncovered the fundamental physical properties that lead to their high mobilities: small effective mass, high sound velocity, high optical phonon frequency, small ratio of Born charge vs. polarizability, and/or weak electron-phonon coupling. The discovered materials as well as the mechanistic insights bring us a step closer to the next-generation electronics/optoelectronics.

Acknowledgements:

This work is supported by Welch Foundation (F-1959-20210327). The calculations were performed on the clusters of Texas Advanced Computing Center (TACC).

References:

[1] S. Jiang, L. Li, Z. Wang, K. F. Mak, and J. Shan, Controlling Magnetism in 2D CrI$_3$ by Electrostatic Doping, Nature Nanotech 13, 549 (2018).
[2] Y. Wang et al., Structural Phase Transition in Monolayer MoTe$_2$ Driven by Electrostatic Doping, Nature 550, 487 (2017).
[3] S. Yu, X. Wu, Y. Wang, X. Guo, and L. Tong, 2D Materials for Optical Modulation: Challenges and Opportunities, Adv. Mater. 29, 1606128 (2017).
[4] D. Akinwande, N. Petrone, and J. Hone, Two-Dimensional Flexible Nanoelectronics, Nat Commun 5, 5678 (2014).
[5] G.-H. Lee et al., Flexible and Transparent MoS$_2$ Field-Effect Transistors on Hexagonal Boron Nitride-Graphene Heterostructures, ACS Nano 7, 7931 (2013).
[6] H.-Y. Chang, S. Yang, J. Lee, L. Tao, W.-S. Hwang, D. Jena, N. Lu, and D. Akinwande, High-Performance, Highly Bendable MoS$_2$ Transistors with High-K Dielectrics for Flexible Low-Power Systems, ACS Nano 7, 5446 (2013).
[7] B. Radisavljevic, A. Radenovic, J. Brivio, V. Giacometti, and A. Kis, Single-Layer MoS$_2$ Transistors, Nature Nanotech 6, 147 (2011).
[8] L. Li, Y. Yu, G. J. Ye, Q. Ge, X. Ou, H. Wu, D. Feng, X. H. Chen, and Y. Zhang, Black Phosphorus Field-Effect Transistors, Nature Nanotech 9, 372 (2014).
[9] D. A. Bandurin et al., *High Electron Mobility, Quantum Hall Effect and Anomalous Optical Response in Atomically Thin InSe*, Nature Nanotech 12, 223 (2017).

[10] S. Haastrup et al., *The Computational 2D Materials Database: High-Throughput Modeling and Discovery of Atomically Thin Crystals*, 2D Mater. 5, 042002 (2018).

[11] M. N. Gjerding et al., *Recent Progress of the Computational 2D Materials Database (C2DB)*, 2D Mater. 8, 044002 (2021).

[12] V. Kuček, C. Drasar, J. Navratil, L. Benes, and P. Lostak, *Optical and Transport Properties of GaGeTe Single Crystals*, Journal of Crystal Growth 380, 72 (2013).

[13] W. Wang, L. Li, Z. Zhang, J. Yang, D. Tang, and T. Zhai, *Ultrathin GaGeTe P-Type Transistors*, Appl. Phys. Lett. 111, 203504 (2017).

[14] L. Cheng, C. Zhang, and Y. Liu, *Why Two-Dimensional Semiconductors Generally Have Low Electron Mobility*, Phys. Rev. Lett. 125, 177701 (2020).

[15] S. Poncé, E. R. Margine, and F. Giustino, *Towards Predictive Many-Body Calculations of Phonon-Limited Carrier Mobilities in Semiconductors*, Phys. Rev. B 97, 121201 (2018).

[16] T. Sohier, D. Campi, N. Marzari, and M. Gibertini, *Mobility of Two-Dimensional Materials from First Principles in an Accurate and Automated Framework*, Phys. Rev. Materials 2, 114010 (2018).

[17] G. Brunin, H. P. C. Miranda, M. Giantomassi, M. Royo, M. Stengel, M. J. Verstraete, X. Gonze, G.-M. Rignanese, and G. Hautier, *Electron-Phonon beyond Fröhlich: Dynamical Quadrupoles in Polar and Covalent Solids*, Phys. Rev. Lett. 125, 136601 (2020).

[18] V. A. Jhalani, J.-J. Zhou, J. Park, C. E. Dreyer, and M. Bernardi, *Piezoelectric Electron-Phonon Interaction from Ab Initio Dynamical Quadrupoles: Impact on Charge Transport in Wurtzite GaN*, Phys. Rev. Lett. 125, 136602 (2020).

[19] S. Poncé, F. Macheda, E. R. Margine, N. Marzari, N. Bonini, and F. Giustino, *First-Principles Predictions of Hall and Drift Mobilities in Semiconductors*, Phys. Rev. Research 3, 043022 (2021).

[20] S. Poncé, W. Li, S. Reichardt, and F. Giustino, *First-Principles Calculations of Charge Carrier Mobility and Conductivity in Bulk Semiconductors and Two-Dimensional Materials*, Rep. Prog. Phys. 83, 036501 (2020).

[21] C. Zhang and Y. Liu, *Phonon-Limited Mobility of 2D Semiconductors: Quadrupole Scattering and Free-Carrier Screening*, arXiv:2207.00110.

[22] L. Cheng, C. Zhang, and Y. Liu, *The Optimal Electronic Structure for High-Mobility 2D Semiconductors: Exceptionally High Hole Mobility in 2D Antimony*, J. Am. Chem. Soc. 141, 16296 (2019).

[23] C. J. Ciccarino, T. Christensen, R. Sundararaman, and P. Narang, *Dynamics and Spin-Valley Locking Effects in Monolayer Transition Metal Dichalcogenides*, Nano Lett. 18, 5709 (2018).

[24] L. Cheng and Y. Liu, *What Limits the Intrinsic Mobility of Electrons and Holes in Two Dimensional Metal Dichalcogenides?*, J. Am. Chem. Soc. 140, 17895 (2018).

[25] T.-H. Liu, J. Zhou, B. Liao, D. J. Singh, and G. Chen, *First-Principles Mode-by-Mode Analysis for Electron-Phonon Scattering Channels and Mean Free Path Spectra in GaAs*, Phys. Rev. B 95, 075206 (2017).

[26] T. Sohier, M. Gibertini, M. Calandra, F. Mauri, and N. Marzari, *Breakdown of Optical Phonons’ Splitting in Two-Dimensional Materials*, Nano Lett. 17, 3758 (2017).

[27] J. Qiao, X. Kong, Z.-X. Hu, F. Yang, and W. Ji, *High-Mobility Transport Anisotropy and Linear
Dichroism in Few-Layer Black Phosphorus, Nat Commun 5, 4475 (2014).
Supplementary Materials: Discover 2D semiconductors with high intrinsic carrier mobility

Chenmu Zhang, Ruoyu Wang, Himani Mishra, Yuanyue Liu*

Texas Materials Institute and Department of Mechanical Engineering,
The University of Texas at Austin, Austin, Texas 78712, USA
Yuanyue.liu@austin.utexas.edu

Supplementary Note S1: Mobility calculation

The carrier mobility $\mu$ for band transport at low electric field can be obtained from the Boltzmann transport theory under momentum relaxation time approximation:

$$\mu_{\alpha\beta} = \frac{q_1}{n_1\Omega} \sum_n \frac{d\Omega}{\Omega_{BZ}} \frac{\partial f_{nk}}{\partial E_{nk}} \tau_{nk} V_{nk,\alpha} V_{nk,\beta}, \quad (S1)$$

where $\alpha$ and $\beta$ are the direction indices, $q_1$ is the charge of carrier, $\Omega$ ($\Omega_{BZ}$) is the area of unit cell (Brillouin zone); $\tau_{nk}$ is the momentum relaxation time for the electronic state with band index $n$ and wavevector $k$, $V_{nk}$ is its group velocity, and $E_{nk}$ is its energy; $f_1$ is the Fermi distribution function, and $n_c$ is the carrier density which is related with $f$ and the electronic band structure through:

$$n_c = \sum_n \int \frac{d\Omega}{\Omega_{BZ}} f_{nk}; \quad n_h = \sum_n \int \frac{d\Omega}{\Omega_{BZ}} (1 - f_{nk}), \quad (S2)$$

where $n_c$ and $n_h$ are the concentrations for electrons and holes respectively. Since we are interested in the intrinsic mobility, we consider a perfect material with the Fermi level at the middle of the band gap and the phonon-induced scattering only. The $\tau_{nk}$ can be calculated as:

$$\frac{1}{\tau_{nk}} = \frac{2\pi}{\hbar} \sum_{nm\nu} \int \frac{d\Omega}{\Omega_{BZ}} | g_{mn}(k, q) |^2 \frac{(f_{nk+q} + n_{\nu}) \delta(E_{nk} - E_{nk+q} + \hbar \omega_{\nu})}{(1 + n_{\nu} - f_{nk+q}) \delta(E_{nk} - E_{nk+q} - \hbar \omega_{\nu})} \left(1 - \frac{\mathbf{v}_{nk} \cdot \mathbf{v}_{nk+q}}{|| \mathbf{v}_{nk} || \mathbf{v}_{nk+q} ||} \right), \quad (S3)$$

where the initial electronic state $nk$ is scattered to the final state $mk+q$ by interacting with a phonon $\nu q$ with frequency $\omega_{\nu q}$ ($\nu$ is the phonon band index and $q$ is the phonon wavevector); $n$ is the Bose distribution; $\mathbf{v}$ is the group velocity vector; $g_{mn}(k, q)$ is the electron-phonon coupling (EPC) matrix element, which can be computed from density functional perturbation theory (DFPT) and interpolated to a dense grid. The calculation details can be found in Note S2.

Supplementary Note S2: Computational details
The first-principles calculations are performed using the Quantum Espresso (QE) Package [1,2] with Optimized Norm-Conserving Vanderbilt (ONCV) pseudopotentials [3–5] and the Perdew-Burke-Ernzerhof (PBE) exchange-correlation functional [6]. We employ fully-relativistic norm-conserving pseudopotentials, which include spin-orbit coupling (SOC) [7]. The kinetic energy cutoff and the k-grid setting are tested for each material and eventually lead to total energy converging within 5 and 1 meV/atom, respectively. The lattice constants and atomic positions are relaxed with force convergence criterion of $10^{-6}$ Ry/Bohr and energy convergence of $10^{-5}$ Ry. The density functional perturbation theory (DFPT) calculations [8] are performed on $12 \times 12$ q grids for 2D materials with hexagonal lattice, or comparable q grids for the other lattices. The calculations of electron-phonon coupling (EPC) and carrier mobility are performed using the EPW code [9,10], where the EPC are interpolated from sparse k/q grids to fine k/q grids. The sparse k grids are selected to be the same as the sparse q grids (for most materials) or multiples of its q grids to ensure that the deviation of Wannier interpolated band structure from the DFT one is less than 3 meV around band edge (< 0.3 eV). The mobility is calculated from uniform k/q grids. For materials with effective mass $> 0.1$, we chose $300 \times 300$ k/q grids while for materials with effective mass $< 0.1$, $600 \times 600$ k/q are used for mobility convergence. A finite broadening of 5 meV is used to evaluate the Dirac delta functions in scattering rates integration and a phonon frequency cutoff of 5 cm$^{-1}$ is applied. The dipole and quadrupole scattering in 2D materials are included for more accurate EPC interpolation [11] and the Coulomb cutoff technique [12] is used to avoid fictitious electrostatic interaction between periodic images. For quadrupole scattering, we use DFPT to compute the macroscopic charge responses to different atomic displacements at several q points around the q=0, and perform second order polynomial fitting to extract quadrupoles. The details and verification of quadrupoles can be found in Ref. [11].

**Supplementary Note S3: High-mobility 2D semiconductor screening**

In practice, we screen the Computational 2D Materials Database (C2DB) database [13,14] through 5 steps as shown in Figure 1a in the main text. (1) First, we identify semiconductors with electronic band gaps within 0.2 and 2 eV. Note that different methods can give different band gaps. Although the PBE functional is less accurate than the HSE/GW method (and tends to underestimate the bandgap), its results are available for more materials in the C2DB database. Thus we use the PBE band gaps at this step. 896 out of around 4000 2D materials from the database fall in such band gap range. (2) Then we excluded those lack of “dynamical stability” and “stiffness stability” as labelled by the database. 541 2D semiconductors survive after this step. (3) For the remaining materials, we calculate their “combined effective mass” $M$ (see Eq. 1 in the main text for the definition) for both electrons and holes. Here we use MoS$_2$, the most common 2D semiconductor so far with n-type conduction, as a reference. Since its electron has a $M$ of 0.85, we use $M<1$ as a criterion to identify the candidates with electron/hole mobility potentially higher than that of MoS$_2$ electron. We find 149 (173) materials with electron (hole) $M$ satisfying this criterion. (4) For those materials, we calculate the $\mu_{up}$ (defined in Eq. 4 in the main text) for both electrons and holes. Considering that the $\mu_{up}$ for MoS$_2$ electron is about
200 cm²V⁻¹s⁻¹, we use $\mu_{\text{up}} > 180$ cm²V⁻¹s⁻¹ as a criterion and then obtain 58 (82) materials with potential high electron (hole) mobility. (5) From these candidates, we further select materials with less than 9 atoms per unit cell and accurately calculate their electron and/or hole mobilities by DFPT + interpolation method [9–11] (See Note 2 for calculation details). In total, we accurately calculated 81 2D semiconductors in total (with 47 electron and 51 hole mobilities). Note that we drop those with more atoms in the unit cell because of the high computational cost. Nevertheless, we provide the full list in the Note 4 for future study.

To verify the $\mu_{\text{up}}$ is indeed the upper limit of carrier mobility, we plotted $\mu_{\text{up}}$ vs. *ab initio* $\mu$ in Figure S1. The solid line indicates $\mu = \mu_{\text{up}}$ and the dashed line indicates $\mu = 1.38 \mu_{\text{up}}$. Indeed, most materials fall below the solid line, which indicates in most cases, $\mu < \mu_{\text{up}}$. Several scatters locate between $\mu = \mu_{\text{up}}$ and $\mu = 1.38 \mu_{\text{up}}$ which might be due to the computational differences between our work and the database. The outliers located above $\mu = 1.38 \mu_{\text{up}}$ is mainly due to their deviations from parabolic band structures, which violate the effective mass approximation used in the Fröhlich and acoustic deformation potential (ADP) models. For instance, the InAs denoted in Figure S1 presents Rashba spin splitting around its conduction band minimum, which leads to ill definition of band edge effective mass and thus its inaccurate $\mu_{\text{up}}$ ($\mu_{\text{up}} \sim 0.5 \mu$ for InAs). Using $\mu = 1.38 \mu_{\text{up}}$ as a criterion, it is safe to say that most of 2D semiconductors with $\mu > 180 \times 1.38 = 250$ cm²V⁻¹s⁻¹ out of $\sim 4000$ from the database are filtered out through our screening process.

![Figure S1](image1)

**Figure S1** *Ab initio* mobility $\mu$ vs. upper limit mobility $\mu_{\text{up}}$ (given by Eq. 4 in the main text). Blue circle indicates electron mobility and red square indicates hole mobility.

**Supplementary Note S4: Details on Fröhlich mobility model**

The more accurate Fröhlich $g$ matrix for 2D materials can be given by:
where $\Omega$ is the area of unit cell; $\kappa$ is the index of the atom in the unit cell, $M$ is the atomic mass, $Z^*$ is the Born effective charge, $\tau$ is the atomic position in unit cell; $jq$ is the optical phonon labeled by mode index $j$ and wavevector $q$, $\omega$ is the phonon frequency, $e$ is the phonon eigenvector; $nk$ is the initial electronic state with band index $n$ and wavevector $k$, $\psi$ is the electronic wavefunction; $a_{2D}$ is the in-plane polarizability of the 2D material. The accurate evaluation of $g^F$ via Equation S4 requires the knowledge of $\psi$, $\omega$ and $e$, involving additional ab initio calculations. In order to efficiently estimate the Fröhlich mobility for high-throughput screening, we make several approximations in computations, where all the input quantities can be extracted from the C2DB database. These approximations include:

(1) We assume $\langle \psi_{nk,q} | e^{iq(\tau-\tau')} | \psi_{nk} \rangle = 1$ since the Fröhlich scattering is the intravalley scattering and thus the $|q|$ is small. (2) The $e_{\kappa,j}(q)$ is approximated by the value when $q \rightarrow 0$, which can be extracted from the database. Here we specify a special direction $q = q_0$ and assume $q \cdot Z^* \cdot e_{\kappa,j}(q) = (q_0 \cdot Z^* \cdot e_{\kappa,j}(q \rightarrow 0)$ for all $q$ around the zone center. (3) We neglect the $q$ dependence of $\omega_{j,q}$ and approximate it by [15]:

$$\omega^*_{j,q} = \omega^*_{j,q}(q \rightarrow \infty) = \omega^*_{j,q}(q = 0) + \frac{e^2}{4\pi\alpha_{2D} \Omega} \left( \sum_{\kappa} q_{j,\kappa} \cdot Z^* \cdot e_{\kappa,j}(q \rightarrow 0) \right)^2 \right).$$

The selection of $\omega^*$ as $q \rightarrow +\infty$ value instead of that at $q=0$ is due to that we found the most involved LO phonons in the backscattering events in carrier transport have relatively large phonon vector, and consequently the $\omega(q \rightarrow +\infty)$ gives better estimation of Fröhlich mobility.

Then we use the $g^F$ from all optical modes to numerically compute the scattering rates $1/\tau_{nk}$ and the Fröhlich mobility $\mu_F$ via Eq. S3 and Eq. S1, respectively, under effective mass approximation.

**Supplementary Note S5: Full list of 2D semiconductors with potentially high mobility**

In the step 4 of screening process, 58 (82) materials with potential high electron (hole) mobility are filtered out. While in step 5, only those with less than 9 atoms in the unit cell are calculated by accurate ab initio method [11]. Although the criterion on number of atoms significantly reduces the computational cost (only 47(51) mobilities need to be calculated), it is likely to miss some high-mobility 2D semiconductors with No. of atoms $\geq 9$. Here we present the full list of filtered 2D semiconductors in step 4, together with all the calculated ab initio mobility. For isotropic 2D semiconductors, one mobility scalar is reported and for anisotropic materials, two $\mu$ along different direction are reported, among which the $\mu$ along direction 1 corresponds to the lower one and the $\mu$ along direction 2 is for the higher one.
| Materials         | PBE bandgap | HSE bandgap | $\mu_{up}$ | $\mu$ along direction 1 | $\mu$ along direction 2 |
|-------------------|-------------|-------------|------------|-------------------------|-------------------------|
| Sb2               | 0.983       | 1.406       | 1.000E+10  | 49.34                   | 1151                    |
| BP                | 0.898       | 1.362       | 4.939E+04  | 2791                    |                         |
| As2               | 1.483       | 2.019       | 7.315E+03  | 55.7                    |                         |
| GeH2              | 0.920       | 1.488       | 2.069E+03  | 1996                    |                         |
| Ga2Ge2Te2         | 0.674       | 1.165       | 2.005E+03  | 137                     |                         |
| Zr2Te2            | 0.452       | 0.652       | 1.673E+03  | 248.3                   |                         |
| P4                | 0.903       | 1.511       | 1.535E+03  | 103.8                   |                         |
| PdSe2             | 0.558       | 0.906       | 8.336E+02  | 50.77                   |                         |
| As4               | 0.835       | 1.324       | 7.925E+02  | 70.74                   |                         |
| Au2Te2            | 0.618       | 1.067       | 7.395E+02  | 50.77                   |                         |
| Co2H2O4           | 0.391       | 2.596       | 5.901E+02  |                         |                         |
| GaN               | 1.877       | 3.090       | 5.773E+02  | 510                     |                         |
| Ge2Te2            | 0.815       | 1.103       | 5.770E+02  | 47.25                   |                         |
| InAs              | 0.691       | 1.234       | 5.409E+02  | 1001                    |                         |
| F2Si2             | 0.666       | 1.492       | 5.326E+02  | 760.2                   |                         |
| Ag2Te2            | 0.477       | 0.756       | 5.244E+02  | 49.96                   |                         |
| Pb2Te2            | 0.445       | 0.878       | 4.901E+02  | 169.8                   |                         |
| InP               | 1.099       | 1.774       | 4.901E+02  | 542.3                   |                         |
| S2Si              | 1.383       | 2.216       | 4.890E+02  | 67.1                    |                         |
| GaP               | 1.583       | 2.369       | 4.124E+02  | 169.8                   |                         |
| Tl2Te2            | 0.294       | 0.621       | 4.082E+02  | 494.5                   |                         |
| Sn2Se2            | 0.400       | 4.012E+02   | 251.7      |                         |                         |
| Tl2Te2            | 0.539       | 2.472       | 3.908E+02  | 176                     |                         |
| Se2Si             | 0.467       | 1.072       | 3.755E+02  | 412.6                   |                         |
| Sn2Te2            | 0.603       | 0.897       | 3.613E+02  | 101.4                   |                         |
| WO2               | 1.340       | 2.097       | 3.593E+02  | 120.3                   |                         |
| Re4S8             | 1.272       | 1.854       | 3.542E+02  | 232.4                   |                         |
| In2O2             | 0.385       | 0.920       | 3.531E+02  | 187.9                   |                         |
| MoO2              | 0.912       | 1.651       | 3.417E+02  | 200                     |                         |
| GaAs              | 1.119       | 1.719       | 3.258E+02  | 812.1                   |                         |
| Zn2Te2            | 0.545       | 1.294       | 3.190E+02  | 308                     |                         |
| MoW3S8            | 1.532       | 2.064       | 2.873E+02  |                         |                         |
| Mo2W2S8           | 1.532       | 2.062       | 2.812E+02  |                         |                         |
| Mo3WS8            | 1.559       | 2.076       | 2.638E+02  |                         |                         |
| Cd2Te2            | 0.633       | 1.331       | 2.509E+02  | 224.7                   |                         |
| Sb2Se3            | 0.421       | 2.357E+02   | 87.55      |                         |                         |
| Os2S4             | 0.530       | 1.387       | 2.345E+02  | 149.5                   |                         |
| Material    | PBE bandgap | HSE bandgap | $\mu_{up}$ | $\mu$ along direction 1 | $\mu$ along direction 2 |
|-------------|-------------|-------------|-----------|-------------------------|-------------------------|
| Sb2         | 0.983       | 1.406       | 1.000E+10 | 2044                    |                         |
| BP          | 0.898       | 1.362       | 1.375E+04 | 1921                    |                         |
| HfI2        | 0.630       | 1.022       | 1.021E+04 | 4782                    |                         |
| ZrI2        | 0.687       | 1.038       | 6.419E+03 | 5138                    |                         |
| WS2         | 1.534       | 2.055       | 4.069E+03 | 919                     |                         |
| P4          | 0.903       | 1.511       | 2.593E+03 | 23.64                   | 480.3                   |
| Ge2H2       | 0.920       | 1.488       | 2.500E+03 | 995.1                   |                         |
| MoW3S8      | 1.532       | 2.064       | 2.000E+03 |                         |                         |
| As2         | 1.483       | 2.019       | 1.622E+03 | 1216                    |                         |
| Mo2W2S8     | 1.532       | 2.062       | 1.556E+03 |                         |                         |
| Mo3WS8      | 1.559       | 2.076       | 1.330E+03 |                         |                         |
| ZrBrI       | 0.774       | 1.174       | 1.325E+03 | 904.5                   |                         |
| WSSe        | 1.402       | 1.911       | 1.282E+03 | 515                     |                         |
| WSe2        | 1.238       | 1.730       | 1.276E+03 | 578                     |                         |
| As4         | 0.835       | 1.324       | 1.203E+03 | 87.8                    | 218.9                   |
| Ti2CO2      | 0.325       | 1.295       | 1.175E+03 | 346.6                   |                         |
| Ir2I2S2     | 0.215       | 0.998       | 1.052E+03 | 23.87                   | 578                     |

Table S1: Bandgap, upper limit of electron mobility $\mu_{up}$ and ab initio electron mobility $\mu$ for 2D semiconductors from step 4 in the screening process.
|        | Value   | Value   | Value   | Value    |
|--------|---------|---------|---------|----------|
| ZrBr2  | 0.828   | 1.276   | 9.962E+02 | 766      |
| MoS2   | 1.580   | 2.087   | 9.802E+02 | 68.9     |
| GaAs   | 1.119   | 1.719   | 9.754E+02 | 16.17    |
| TiI2   | 0.596   | 0.828   | 9.748E+02 | 884      |
| TiBr2  | 0.758   | 1.166   | 9.384E+02 | 752.7    |
| Ir2Br2O2 | 0.295 | 1.342   | 8.506E+02 | 22.3     |
| Ir2Cl2S2 | 0.355 | 1.201   | 8.469E+02 | 22.75    |
| CrW3S8 | 1.126   |         | 8.379E+02 |           |
| HfBrI  | 0.713   | 1.157   | 8.057E+02 | 805.4    |
| InAs   | 0.691   | 1.234   | 7.210E+02 | 372.2    |
| Mo2W2Se8 | 1.270 | 1.749   | 7.048E+02 |           |
| Mo3WSe8 | 1.300 | 1.785   | 7.014E+02 |           |
| Ir2Br2S2 | 0.344 | 1.178   | 6.848E+02 | 23.46    |
| TiBrI  | 0.679   | 1.037   | 6.791E+02 | 451.2    |
| ZrBrCl | 0.912   | 1.397   | 6.628E+02 | 476.4    |
| CrW3Se8 | 0.882  | 1.333   | 6.428E+02 |           |
| Al2O2  | 1.315   | 1.849   | 6.285E+02 | 258      |
| WTe2   | 0.727   | 1.141   | 5.748E+02 | 348.6    |
| ZrCl2  | 0.981   | 1.500   | 5.655E+02 | 370.3    |
| ZrClI  | 0.877   | 1.317   | 5.550E+02 | 337.4    |
| CrMo3S8 | 1.184  |         | 5.371E+02 |           |
| Co2H2O4 | 0.391  | 2.596   | 5.287E+02 |           |
| Ge2Te2 | 0.815   | 1.103   | 5.024E+02 | 134.5    |
| Hf2CO2 | 0.938   | 1.681   | 4.934E+02 | 190      |
| Pb2Te2 | 0.445   | 0.878   | 4.876E+02 | 84.3     |
| Zr2CO2 | 0.950   | 1.729   | 4.775E+02 | 117.9    |
| Cr2W2S8 | 0.941  | 1.442   | 4.682E+02 |           |
| F2Si2  | 0.666   | 1.492   | 4.575E+02 | 75.53    |
| HfBr2  | 0.732   | 1.226   | 4.393E+02 | 588      |
| Mo2Cl6 | 0.507   | 0.815   | 4.173E+02 | 78.04    |
| MoSe2  | 1.453   | 1.949   | 4.133E+02 | 211.4    |
| WSeTe  | 1.037   | 1.493   | 4.101E+02 |           |
| S2Si   | 1.383   | 2.216   | 3.960E+02 | 30.74    |
| MoSe2  | 1.321   | 1.804   | 3.932E+02 | 146.9    |
| HfCl2  | 0.814   | 1.295   | 3.815E+02 | 481.8    |
| MoW3Te8 | 0.804  | 1.220   | 3.788E+02 |           |
| Cr3MoTe8 | 0.467 | 0.893   | 3.658E+02 |           |
| Se2Si  | 0.467   | 1.072   | 3.581E+02 | 55.26    |
| PtTe2  | 0.303   | 0.599   | 3.536E+02 | 337.1    |
| Ir2Cl2O2 | 0.444 | 1.566   | 3.401E+02 | 24.57    |
| Cr3MoS8 | 0.899  |         | 3.346E+02 |           |
| W2Cl6  | 0.706   | 1.165   | 3.275E+02 |           |
| CrW3Te8 | 0.543  | 0.937   | 3.133E+02 |           |
| Compound       | μ<sub>up</sub> | ε<sub>1</sub> | ε<sub>2</sub> | μ | E<sub>g</sub> |
|---------------|--------------|-------------|-------------|---|---------|
| HfBrCl        | 0.832        | 1.361       | 3.116E+02   | 239.5 |
| Au<sub>2</sub>Te<sub>2</sub> | 0.618        | 1.067       | 3.080E+02   | 235.5 | 248 |
| Mo<sub>2</sub>W<sub>2</sub>Te<sub>8</sub> | 0.862        | 1.279       | 3.025E+02   |          |
| Mo<sub>3</sub>WTe<sub>8</sub> | 0.900        | 1.326       | 2.930E+02   |          |
| GeTe          | 1.474        | 1.994       | 2.820E+02   | 171.6 |
| HfCl<sub>2</sub> | 0.908        | 1.471       | 2.757E+02   | 277.2 |
| Cr<sub>2</sub>W<sub>2</sub>Se<sub>8</sub> | 0.759        | 1.203       | 2.687E+02   |          |
| Au<sub>2</sub>MoSe<sub>4</sub> | 0.592        | 1.247       | 2.610E+02   |          |
| Cr<sub>3</sub>WSe<sub>8</sub> | 0.677        |             | 2.478E+02   |          |
| Sn<sub>2</sub>Te<sub>2</sub> | 0.603        | 0.897       | 2.134E+02   | 135.2 | 237.6 |
| Zr<sub>2</sub>Se<sub>6</sub> | 0.410        | 1.031       | 2.122E+02   |          |
| Cu<sub>2</sub>Se<sub>2</sub> | 0.447        | 1.116       | 2.020E+02   |          |
| Ag<sub>2</sub>Te<sub>2</sub> | 0.477        | 0.756       | 1.994E+02   |          |
| Zn<sub>2</sub>H<sub>8</sub>N<sub>4</sub>Te<sub>2</sub> | 1.822        | 2.567       | 1.966E+02   |          |
| Cr<sub>2</sub>W<sub>2</sub>Te<sub>8</sub> | 0.497        | 0.876       | 1.946E+02   |          |
| Ti<sub>2</sub>Br<sub>2</sub>N<sub>2</sub> | 0.621        | 2.084       | 1.933E+02   | 89.39 | 155.5 |
| Cr<sub>2</sub>Mo<sub>2</sub>Se<sub>8</sub> | 0.819        |             | 1.911E+02   |          |
| Al<sub>2</sub>Si<sub>2</sub>Te<sub>6</sub> | 1.115        | 1.673       | 1.898E+02   |          |
| TiS<sub>6</sub> | 0.295        | 1.452       | 1.897E+02   |          |
| Cr<sub>3</sub>WTe<sub>8</sub> | 0.443        | 0.841       | 1.882E+02   |          |
| MoTe<sub>2</sub> | 0.932        | 1.370       | 1.882E+02   | 81.3  |

**Table S2**: Bandgap, upper limit of hole mobility $\mu_{up}$ and *ab initio* hole mobility $\mu$ for 2D semiconductors from step 4 in the screening process.

**Supplementary Note S6**: Conductivity vs. bandgap
**Figure S2** Conductivity vs. band gap (HSE) for various 2D semiconductors. The electron/hole concentrations are fixed at $10^{13}$ cm$^{-2}$. Note that the free-carrier screening is not considered here, and the scattering rates are assumed to be same as the intrinsic ones.

**Supplementary Note S7: Additional materials list from element substitution**

From *ab initio* mobility calculations, we found 11 2D semiconductors with electron or hole mobility $> 1000$ cm$^2$V$^{-1}$s$^{-1}$, falling into 5 groups of crystal structure (Fig. 1b). To extend the research, we substitute the elements in these 11 high-mobility materials while keeping their atomic structure unchanged. From each parent material, we obtained a series of new materials not present in the C2DB database, and then calculate their properties for further screening. Here we focus on bandgap, combined effective mass, both of which can be obtained from electronic structure, and dynamical stability (i.e., no negative phonon frequency). For those survive above 3 criteria, we calculate the their corresponding electron/hole mobilities. The additional materials list and screening process are shown in Table S3. Note that here we used a slightly stricter $M<0.5$ criterion than previous screening for efficiency.
| Materials      | Parent materials | 0.2<bandgap<2 eV? | M<0.5? | Dynamical stability? | Electron(hole) Mobility |
|---------------|------------------|-------------------|--------|----------------------|------------------------|
| Ga2Ge2S2      | Ga2Ge2Te2        | No                |        |                      |                        |
| Ga2Sn2Te2     | Ga2Ge2Te2        | Yes               | No     |                      |                        |
| In2Ge2Te2     | Ga2Ge2Te2        | No                |        |                      |                        |
| In2Ge2Te2     | Ga2Ge2Te2        | No                |        |                      |                        |
| Ga2Si2S2      | Ga2Ge2Te2        | Yes               | Yes    | Yes                  | 178                    |
| Ga2Si2Se2     | Ga2Ge2Te2        | Yes               | Yes    | Yes                  | 260                    |
| Ga2Si2Te2     | Ga2Ge2Te2        | Yes               | No     |                      |                        |
| Al2Si2S2      | Ga2Ge2Te2        | Yes               | No     |                      |                        |
| Al2Si2Se2     | Ga2Ge2Te2        | Yes               | No     |                      |                        |
| Al2Ge2S2      | Ga2Ge2Te2        | Yes               | No     |                      |                        |
| Al2Ge2Se2     | Ga2Ge2Te2        | Yes               | Yes    | Yes                  | 421                    |
| Al2Ge2Te2     | Ga2Ge2Te2        | Yes               | Yes    | Yes                  | 2023                   |
| GaSb          | BSb              | Yes               | Yes    | Yes                  | 1809(1080)             |
| InSb          | BSb              | No                |        |                      |                        |
| Bi            | BSb              | Yes               | Yes    | Yes                  | 523(548)               |
| AlBi          | BSb              | Yes               | Yes    | Yes                  | 2835(3446)             |
| GaBi          | BSb              | No                |        |                      |                        |
| InBi          | BSb              | No                |        |                      |                        |
| Ge2Cl2        | Ge2H2            | No                |        |                      |                        |
| Ge2Br2        | Ge2H2            | No                |        |                      |                        |
| Ge2I2         | Ge2H2            | Yes               | No     |                      |                        |
| Si2Cl2        | Ge2H2            | Yes               | Yes    | Yes                  | 198(406)               |
| Si2Br2        | Ge2H2            | Yes               | Yes    | Yes                  | 406                    |
| Si2I2         | Ge2H2            | Yes               | Yes    | Yes                  |                        |

**Table S3** Additional materials with potentially high mobility from element substitution.

**Supplementary Note S8: Ratio of the (maximum) Born charge to the in-plane polarizability**

In Figure 4, we compare the LO $|g|^2$ for BSb hole, ZrI$_2$ hole, Ga$_2$Ge$_2$Te$_2$ electron and MoS$_2$ electron and account for the difference of LO $|g|^2$ by $R_{BP}$, which is defined as the ratio of the maximum born charge to the in-plane polarizability. This is due to from Eq. 2, larger Born effective charges and smaller in-plane polarizability both lead to higher LO EPC $g$ matrix and stronger Fröhlich scattering. Here we use the maximum absolute value of Born effect charge elements to represent the Born effective charges matrix. In Table S4, we list the maximum Born charges, in-plane polarizabilities and $R_{BP}$ values for BSb, ZrI$_2$, Ga$_2$Ge$_2$Te$_2$ and MoS$_2$, in descending order of $R_{BP}$.

| Materials | Maximum Born charge (e) | In-plane polarizability (Å) | $R_{BP}$ |
|-----------|-------------------------|----------------------------|----------|
| BSb       | 6.0                     | 11.8                       | 0.51     |
Table S4 Maximum Born charges, in-plane polarizabilities and $R_B/P$ values for BSb, ZrI$_2$, Ga$_2$Ge$_2$Tr$_2$ and MoS$_2$.

| Material   | $Ga^2$ | $Ge^2$ | $Te^2$ | $MoS^2$ | $ZrI^2$ |
|------------|--------|--------|--------|---------|---------|
|            | 2.4    | 10.6   | 0.23   | 0.5     | 0.0012  |

**Supplementary Note S9: Drude effective mass and Drude scattering rate**

To understand the high mobilities, we calculate “Drude effective mass” ($\mu^*$) and “Drude scattering rate” ($1/\tau$). They are defined by re-writing Equation S1 in a form similar to the Drude model:

$$\mu = \frac{|q|}{\tau}$$  \hspace{1cm} (S7)

where

$$\frac{1}{\tau} = \frac{1}{2n_\Omega} \sum_{\nu} \int \frac{d\mathbf{k}}{\Omega_{BZ}} \frac{\partial f_{\mathbf{k},\nu}}{\partial E_{\mathbf{k},\nu}},$$  \hspace{1cm} (S8)

$$\tau = \frac{\mu\tau^*}{|q|}.$$

The $\tau^*$ is fully determined by the electronic structure and its occupation, while the information about phonons and the EPCs are wrapped in $1/\tau$. Fig. 3a shows the $1/\tau$ vs. $\tau^*$ for the materials with e/h mobility $> 1000$ cm$^2$V$^{-1}$s$^{-1}$, as well as those for MoS$_2$ for comparison.

**Supplementary Note S10: Density of scatterings**

The “density of scatterings” $D^S$ is used to understand the differences in the mode-resolved scattering. The $D^S$ has a format similar to the scattering rate expression in Eq. S3:

$$D_{\nu,nk}^S = \sum_n \int_{BZ} \frac{d\mathbf{q}}{\Omega_{BZ}} ((f_{\mathbf{nk}+\mathbf{q}} + n_{\mathbf{vq}}) \delta(E_{\mathbf{nk}} - E_{\mathbf{nk}+\mathbf{q}} + \hbar\omega_{\mathbf{vq}})$$

$$+ (1 + n_{\mathbf{vq}} - f_{\mathbf{nk}+\mathbf{q}}) \delta(E_{\mathbf{nk}} - E_{\mathbf{nk}+\mathbf{q}} - \hbar\omega_{\mathbf{vq}}) \frac{\nu_{\mathbf{nk}} \cdot \nu_{\mathbf{nk}+\mathbf{q}}}{|\nu_{\mathbf{nk}}| \cdot |\nu_{\mathbf{nk}+\mathbf{q}}|})$$  \hspace{1cm} (S9)

except the $|g|^2$ is not contained and the phonon modes are not summed. The $D^S$ is determined by the “match” between electron and phonon structures (as described by the delta functions) as well as their occupations, without involving the EPCs. It characterizes the density of scattering events for an initial state $nk$ and a specific phonon mode $\nu$.

**Supplementary Note S11: Mobility vs. several basic physical properties**
Figure S3 Mobility vs. several basic and easy-to-calculate physical properties: combined effective mass (a), the ratio of maximum in-plane Born charge to in-plane polarizability (b), longitudinal sound velocity (c), and LO phonon frequency (d). The high-mobility (> 1000 cm²V⁻¹s⁻¹) 2D semiconductors are highlighted by red (for electron) and blue (for hole).

References:

[1] P. Giannozzi et al., QUANTUM ESPRESSO: A Modular and Open-Source Software Project for Quantum Simulations of Materials, J. Phys.: Condens. Matter 21, 395502 (2009).
[2] P. Giannozzi et al., Advanced Capabilities for Materials Modelling with Quantum ESPRESSO, J. Phys.: Condens. Matter 29, 465901 (2017).
[3] M. Schlipf and F. Gygi, Optimization Algorithm for the Generation of ONCV Pseudopotentials, Computer Physics Communications 196, 36 (2015).
[4] D. R. Hamann, Optimized Norm-Conserving Vanderbilt Pseudopotentials, Phys. Rev. B 88, 085117 (2013).
[5] M. J. van Setten, M. Giantomassi, E. Bousquet, M. J. Verstraete, D. R. Hamann, X. Gonze, and G.-M. Rignanese, The PseudoDojo: Training and Grading a 85 Element Optimized Norm-Conserving Pseudopotential Table, Computer Physics Communications 226, 39 (2018).
[6] J. P. Perdew, K. Burke, and M. Ernzerhof, Generalized Gradient Approximation Made Simple, Phys. Rev. Lett. 77, 3865 (1996).
[7] P. Scherpelz, M. Govoni, I. Hamada, and G. Galli, *Implementation and Validation of Fully Relativistic GW Calculations: Spin–Orbit Coupling in Molecules, Nanocrystals, and Solids*, J. Chem. Theory Comput. 12, 3523 (2016).

[8] P. Giannozzi, S. de Gironcoli, P. Pavone, and S. Baroni, *Ab Initio Calculation of Phonon Dispersions in Semiconductors*, Phys. Rev. B 43, 7231 (1991).

[9] J. Noffsinger, F. Giustino, B. D. Malone, C.-H. Park, S. G. Louie, and M. L. Cohen, *EPW: A Program for Calculating the Electron–Phonon Coupling Using Maximally Localized Wannier Functions*, Computer Physics Communications 181, 2140 (2010).

[10] S. Poncé, E. R. Margine, C. Verdi, and F. Giustino, *EPW: Electron–Phonon Coupling, Transport and Superconducting Properties Using Maximally Localized Wannier Functions*, Computer Physics Communications 209, 116 (2016).

[11] C. Zhang and Y. Liu, *Phonon-Limited Mobility of 2D Semiconductors: Quadrupole Scattering and Free-Carrier Screening*, arXiv:2207.00110.

[12] T. Sohier, M. Calandra, and F. Mauri, *Density Functional Perturbation Theory for Gated Two-Dimensional Heterostructures: Theoretical Developments and Application to Flexural Phonons in Graphene*, Phys. Rev. B 96, 075448 (2017).

[13] S. Haastrup et al., *The Computational 2D Materials Database: High-Throughput Modeling and Discovery of Atomically Thin Crystals*, 2D Mater. 5, 042002 (2018).

[14] M. N. Gjerding et al., *Recent Progress of the Computational 2D Materials Database (C2DB)*, 2D Mater. 8, 044002 (2021).

[15] T. Sohier, M. Gibertini, M. Calandra, F. Mauri, and N. Marzari, *Breakdown of Optical Phonons’ Splitting in Two-Dimensional Materials*, Nano Lett. 17, 3758 (2017).