Van der Waals Interface Transistors as Light Sources with Bias Tunable Spectrum

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(Dated: January 5, 2022)

Light-emitting electronic devices are ubiquitous in key areas of current technology, such as data communications, solid-state lighting, displays, and optical interconnects. Controlling the spectrum of the emitted light electrically, by simply acting on the device bias conditions, is an important goal with potential technological repercussions. However, identifying a material platform enabling broad electrical tuning of the spectrum of electroluminescent devices is difficult. Here, we propose light-emitting field-effect transistors based on van der Waals interfaces of atomically thin semiconductors as a promising class of devices to achieve this goal. We demonstrate that large spectral changes in room-temperature electroluminescence can be controlled both at the device assembly stage—by suitably selecting the material forming the interfaces—and on-chip, by changing the bias to modify the device operation point. As the physical mechanism responsible for light emission is robust and does not depend on details of the interfaces, these structures are compatible with simple large areas device production methods.

Light-emitting field-effect transistors (LEFETs) are three-terminal devices that allow switching of both the electrical conductance and light emission.1–5 They rely on semiconductors that support ambipolar transport to inject simultaneously in the transistor channel electrons and holes,6 whose radiative recombination is the origin of the emitted light.7,8 Past research on LEFETs has concentrated on organic semiconductors, which have suitable properties for their realization.4,5,9–11 Ionic gated LEFETs based on 2D semiconductors (Figure 1a and b and Supplementary Section S2) are a recently discovered alternative that offer potential advantages, such as higher and well-balanced electron and hole mobilities, as well as low-bias operation.12–16 Efficient LEFETs, however, require the use of 2D semiconductors with a direct bandgap, whose paucity limits the possibility to tune the spectrum of the emitted light. Van der Waals (vdW) interfaces formed by atomically thin semiconducting materials provide a strategy to address this issue because the wavelength of light emitted by interlayer transitions (electrons hosted in one layer recombining with holes hosted in the other, Figure 1c) can be engineered by selecting constituent materials with an appropriate band alignment.17–21

Here, we demonstrate experimentally LEFETs realized on vdW interfaces, and show that they can be operated as electrically tunable light sources. As compared to LEFETs based on individual monolayers, devices fabricated on vdW interfaces potentially offer more functionality. The electronic structure of the individual layers, for instance, is often only minorly affected by the interface formation, so that a rich set of electronic levels—i.e., the bands of the two materials, including the sub-bands originating from quantum confinement—is present (Figure 1d).22–26 If properly populated by acting on the device operation point (i.e., the applied source-drain and gate voltages, $V_{SD}$ and $V_G$, respectively), these levels may enable the energy of the emitted light to be tuned. Additionally, the electric field perpendicular to the transistor channel creates a potential difference between the layers forming the interface, which shifts the energy of the recombining electrons and holes.27,28 The wavelength of light generated by interlayer transitions is expected to shift accordingly, providing another route to tune the emission spectrum by acting on the device operation point. These ideas disclose possible mechanisms to operate LEFETs based on vdW interfaces as electrically tunable light sources. However, neither their validity nor

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Figure 1. LEFETs based on van der Waals interfaces. a. Schematics of an ionic gated field-effect transistor, with the source-drain (S-D) contacts connected to a semiconducting layer and the gate electrode, all in contact with an ionic liquid. The zoom in on the channel regions shows that upon the application of a gate voltage charge is accumulated on the semiconductor (see Supplementary Section S2 for details). b and c. Electron-hole recombination in the channel of a LEFET operated in the ambipolar injection regime (with electrons and holes injected at opposite contacts) for a device based on a individual 2D material (b) and on a vdW interface (c). In the latter case, light is emitted by the recombination of electrons and holes hosted in different layers, which offers new opportunity to control its spectrum (see main text). d. The type-II band alignment between InSe and TMDs around the Γ-point enables k-direct interlayer radiative transitions between the two band edges (as indicated by the orange arrow). Higher energy bands originating from the quantum confinement of charge carriers in the two layers (as indicated by the thin lines) can lead to radiative transitions with different energy. e. Optical micrograph of a device used in this work, based on 2L WS$_2$ and 4L InSe (the contours of the layers are marked by the blue and white lines). The vdW interface is in contact with the liquid through an opening in the PMMA layer covering the entire sample. The scale bar is 2 μm. f. Peak in the PL spectrum of a 5L InSe crystal encapsulated between two thicker hBN layers: the narrow width indicates the high quality of the material.

Figure 2. Transistor characteristics of a 2L WS$_2$/4L InSe device. a. Room-temperature transfer curve ($I_{SD}$-vs-$V_G$ at fixed $V_{SD} = 50$ mV) of a 2L-WS$_2$/4L-InSe. b. The high device quality is evidenced by the low subthreshold swings for electron and hole transport, whose values (115 mV/decade and 90 mV/decade, respectively, for this specific transistor) are close to the ultimate limit of 60 mV/decade. c and d. Output characteristics ($I_{SD}$ as function of $V_{SD}$) of the same device for different negative and positive gate biases, respectively. Linear, saturation, and ambipolar injection regimes can be clearly identified upon increasing the magnitude of $V_{SD}$, as described in the main text (see also Supplementary Section S2).

the potential of LEFETs based on vdW interfaces could be assessed so far, because no such a device has been reported to date.

Our work relies on devices made of bilayers (2L) of semiconducting transition metal dichalcogenides (TMDs; we use WS$_2$ and MoS$_2$) and InSe multilayers (Figure 1e), to form vdW interfaces that belong to a recently identified class exhibiting robust radiative interlayer transitions (e.g., transitions that are radiative irrespective of the lattice structure of the constituent materials or of their relative orientation)\textsuperscript{24,29,30} The robustness originates from having the conduction and valence band extrema in the two layers at $k = 0$, i.e., at the Γ-point of the Brillouin zone (Figure 1d), which is why we refer to these systems as to Γ-Γ interfaces. It is important because it facilitates the device assembly, and makes it compatible with simple large-area production techniques.\textsuperscript{31,32} Indeed, we find that all our LEFETs exhibit electroluminescence, with a wavelength that can be engineered by selecting the constituent layers, and with a spectrum that can be tuned by acting on the device operation point. Contrary to earlier studies of Γ-Γ interfaces\textsuperscript{30} – in which photoluminescence (PL) was only observed at cryogenic temperatures – electroluminescence (EL) is already present at room temperature, a key finding when assessing the technological potential of these devices.
Figure 3. **Electroluminescence from a Γ-Γ' interface.**

a. Images of the channel of a transistor based on a 2L-WS₂/4L-InSe heterostructure taken with an optical microscope (top) and with the camera of our spectrometer (bottom). The bottom image, taken with the device biased at the onset of the ambipolar injection regime, shows a bright spot due to EL. The scale bars are 1 μm.

b. Device output curve measured at, V₆₅ = +0.5 V (electron accumulation).

c. False-color plot of the EL intensity measured by the spectrometer, as a function of photon energy and applied V₆₅, showing a peak centered around 1.25 eV that emerges at V₆₅ = +1.9 V, corresponding to the onset of ambipolar injection (see b).

d. Individual EL spectra at selected V₆₅ values extracted from c: in this bias range the spectrum remains unchanged, and the intensity increases following the increase in source-drain current.

e-g. Data analogous to those of panels b-d are shown for V₆₅ = -2.2 V (hole accumulation), demonstrating that the presence of EL is robust and that at sufficiently low bias the spectrum is virtually identical for electron and hole accumulation.

h. The peak in the EL spectrum (acquired with V₆₅ = +2.2 V and V₆₅ = +0.5 V) is red-shifted relative to the PL emission energies of the layers forming the interface (purple: 4L InSe; blue: 2L WS₂; for InSe, PL data are taken at 5 K, because no PL is observed at room temperature), as expected from an interlayer Γ-Γ' transition (see i).

i-j. Comparison between the normalized EL (thick red line) and PL (thin red line) emission spectra of the interface. The energy difference originates from having to measure PL at cryogenic temperatures (T = 5 K), since no PL is observed at room temperature.

**Device fabrication and characterization**

Ionic gated LEFETs (see Figure 1a-c and Supplementary Section S2) based on 2L-TMD/InSe vdW interfaces are realized using techniques commonly employed for the assembly of structures based on 2D materials.³³ TMD bilayers and InSe multilayers are exfoliated from bulk crystals onto Si/SiO₂ substrates. Heterostructures are formed by picking up layers one after the other, and transferring the resulting interface onto a fresh Si/SiO₂ substrate, with the TMD layer covering the InSe one and effectively encapsulating it (which is important in view of the non-perfect stability of InSe in ambient; the interface assembly process is carried out in the controlled atmosphere of a glove box). Source and drain contacts, as well as a large pad acting as gate electrode, are defined by means of electron beam lithography, electron-beam evaporation of a Pt-Au film (5/30 nm) and lift off. Subsequently, a window in PMMA is patterned to define the region where the ionic liquid contacts the interface. The liquid is applied as a final step, prior to inserting the devices in a vacuum chamber with optical access (see Supplementary Section S1 for more details).

The WS₂ and MoS₂ crystals used for exfoliation are purchased from HQ Graphene. InSe is a less commonly
Figure 4. **LEFET based on a MoS\textsubscript{2}/InSe interface.** a and b. Transfer and output characteristics of a 2L-MoS\textsubscript{2}/5L-InSe transistor. c. EL spectra of the device biased near the onset of ambipolar injection, for electron and hole accumulation (the grey and blue curves are measured respectively at $V_G = -0.7$ V and $V_{SD} = +2.1$ V, and at $V_G = -2.7$ V and $V_{SD} = -2.3$ V), showing a peak around 1.3 eV independently of the applied gate voltage. d. Dependence of EL intensity on photon energy and $V_{SD}$, measured at $V_G = -0.7$ V, showing that near the onset of ambipolar transport the spectrum is independent of $V_{SD}$. e. Also in this case, the EL spectrum (thick orange line) is red-shifted as compared to the PL emission energy of the layers forming the interface (purple: 4L InSe; green: 2L MoS\textsubscript{2}), as expected for an interlayer transition. f. EL spectrum of LEFETs realized using four different interfaces, based on two different 2L TMDs (blue: MoS\textsubscript{2}; red: WS\textsubscript{2}) and three different thicknesses of the InSe layer (3L, 4L, and 5L, as indicated in the figure), showing a dense coverage of part of the visible spectrum (a broader range of photon energy can be spanned using other semiconducting TMD compounds).

Figure 2a shows the room-temperature transfer curve (source-drain current $I_{SD}$ versus gate voltage $V_G$ at fixed source-drain voltage $V_{SD} = 50$ mV) of a device realized on a 2L-WS\textsubscript{2}/4L-InSe interface. The behavior is typical of ambipolar transistors with current mediated by holes and electrons flowing for sufficiently large negative and positive $V_G$, respectively.\textsuperscript{10,12,37–42} Accumulation of electrons and holes leads to comparable current levels, confirming that transport is well-balanced and that residual defects in InSe do not prevent high-quality device operation. When plotted in logarithmic scale (Figure 2b) the data allow determining the subthreshold swing $S = \ln 10 \frac{dV_G}{d(I_{SD})}$, equal to $S = 115$ and 90 mV/decade near the threshold for hole and electron conduction, respectively (other devices exhibit even closer to the ultimate room-temperature limit of 60 mV/decade).\textsuperscript{43} The output curves ($I_{SD}$-vs-$V_{SD}$) shown in Figure 2c and 2d also exhibit the expected behavior. Upon increasing $V_{SD}$, $I_{SD}$ increases linearly at first, then saturates, and eventually exhibits a very steep increase, when entering the ambipolar injection regime. This regime—in which electrons and holes are injected at opposite contacts—is the one of interest for LEFET operation (see Supplementary Section S2), and can be reached irrespective of the polarity of the applied gate voltage.

EL is expected to occur concomitantly with ambipolar injection, with light emission starting at one of the contacts and shifting into the channel as $V_{SD}$ is further increased.\textsuperscript{4} This is indeed what we observe (see Figure 3a as well as Supplementary Figure S2 and accompanying discussion in Supplementary Section S2). The light emitted by the LEFET is collected by a microscope objective and fed into a spectrometer. The spectral analysis performed on data measured at fixed $V_G$, by increasing $V_{SD}$ past the onset of the ambipolar injection regime ($V_{SD} > +1.9$ V in Figure 3b for $V_G > 0$ and $V_{SD} < -2.2$ V in Figure 3e for $V_G < 0$), is shown in Figures 3c and 3d for $V_G > 0$ V, and in Figures 3f and 3g for $V_G < 0$ V. As the current increases exponentially rapidly, we initially limit the maximum applied $V_{SD}$ to
is measured at $T = 5$ K, since no signal is observed at room temperature) and 2L-WSe$_2$ (blue line). The energy of the EL peak is considerably lower than the recombination energy in either 4L-InSe or 2L-WSe$_2$, as expected for an interlayer transition.\textsuperscript{30} The energy of the room-temperature EL signal (Figure 3j, thick line) matches that of low-temperature interlayer transitions seen in PL (Figure 3j, thin line) due to electrons in InSe recombining with holes in WS$_2$, if we take into account that the TMD gap typically increases by approximately 100 meV upon cooling from 300 to 5 K.\textsuperscript{44,45} The measurements, therefore, confirm that our LEFET operates as anticipated, with electrons injected in the InSe layer and holes in the WS$_2$ one recombining via an interlayer transition. Finding that this transition results in EL even at room temperature is a positive, unexpected surprise.

Devices based on other $\Gamma$-$\Gamma$ vdW interfaces should exhibit all key properties of 2L-WSe$_2$/4L-InSe LEFETs. We verify that this is indeed the case using transistors realized on interfaces of 2L-MoS$_2$ (instead of 2L-WSe$_2$), and 3L-, 4L-, and 5L-InSe. Without going through all details, the data show the occurrence of ambipolar transport (Figure 4a) and of the ambipolar injection regime past saturation (Figure 4b). Upon entering the ambipolar injection regime, EL is observed resulting in a line at 1.3 eV (for 2L-MoS$_2$/5L-InSe) independently of $V_{SD}$ (see Figure 4c and 4d), i.e., an energy lower than that of the transitions in the constituent materials (see Figure 4e). Figure 4f overviews the results obtained, by plotting together the room-temperature EL spectrum of LEFETs fabricated on all different vdW interfaces, and shows that combining different 2D materials indeed allows a dense coverage of part of the near-infrared and visible spectral range. Selecting multilayers of different thicknesses or having different compositions (e.g., MoSe$_2$ or MoTe$_2$) would further broaden the accessible spectrum, both on the higher and lower end.\textsuperscript{24,29,30}

**Electrically tunable EL spectrum in vdW interface LEFET**

Having established that LEFETs based on $\Gamma$-$\Gamma$ interfaces provide a robust platform to generate room-temperature EL, we test whether the light spectrum can be controlled by varying the device operation point. Figure 5 illustrates the evolution of the spectrum of the light emitted by a 2L-WSe$_2$/4L-InSe (Figure 5a-c) and by a 2L-MoS$_2$/5L-InSe (Figure 5d-f) LEFETs, upon pushing the source-drain bias $V_{SD}$ to reach deeper in the ambipolar transport regime. The green rectangle in Figure 5a delimits the $V_{SD}$ interval discussed earlier, and the corresponding part of the spectrum in Figure 5c (also delimited by a green rectangle) shows emission from the interlayer transition just above 1.2 eV, in agreement

![Figure 5. Bias tunable light emission from vdW interface LEFETs. a. Output characteristics of the 2L-WSe$_2$/4L-InSe device, whose data are shown in Figure 3 ($V_G = +0.2$ V). The colored dashed lines delimit the low-bias (green line) and high-bias (red line) regime, which exhibit different EL spectral properties. b. Color plot of the EL spectrum as a function of photon energy and $V_{SD}$. In the low-bias regime (region inside the green rectangle) EL exhibit a single peak, due to a $\Gamma$-$\Gamma$ interlayer transition from the bottom of the InSe conduction band to the top of the WS$_2$ valence band (see Figure 3). In this regime the spectrum is independent of bias. In the high-bias regime (region inside the red rectangle), the EL spectrum evolves upon increasing $V_{SD}$, showing that the LEFET acts a light source with bias-tunable spectrum. c. Individual EL emission spectra measured in the high-bias regime, upon varying $V_{SD}$ from 2.3 to 2.5 V. d-f. Same measurements as those shown in panels d-f performed on a LEFET realized on a 2L-MoS$_2$/5L-InSe device (data taken at $V_G = -0.7$ V). The data illustrate that the evolution of the EL spectrum observed in this MoS$_2$/InSe LEFET is fully analogous that observed in the WS$_2$/InSe LEFET, showing the robustness of the device operation.
is fixed to +2.4 V). The data show that varying $V_{SD}$ of the intensity of the light emitted by a 2L-WS$_2$/4L-InSe LEFET device as function of photon energy and gate voltage ($V_{SD}$ is fixed to +2.4 V). The spectrum is peaked at 1.25 eV, i.e., the characteristic energy of the interlayer transition between the bottom of the InSe conduction band and the top of the WS$_2$ valence band; for $V_G < 0.2$ V a broader peak centered around 1.4 eV and shifting with varying $V_G$ is observed. Individual horizontal cuts of the color plot shown in a, for $V_G$ varying from -0.1 V to +0.7 V in 0.1 V steps. c and d. Same measurements as in a and b with $V_{SD}$ fixed at +3.12 V. The individual spectral cuts range from $V_G = +0.2$ V to +1.1 V in 0.1 V steps.

with the data shown in Figure 3b and 3c. When larger $V_{SD}$ is applied, corresponding to the interval in the rectangle delimited by the red line in Figure 5a, the spectrum evolves. Additional transitions appear, visible in Figure 5b in the region delimited by the red rectangle, as well as in Figure 5c, which shows the spectrum of the emitted light at specific values of $V_{SD}$. A qualitatively identical behavior is observed in LEFETs based on 2L-MoS$_2$/5L-InSe, with the regime of lower and higher $V_{SD}$ highlighted by red and green rectangles in Figure 5d and the corresponding spectra shown in Figure 5e and 5f.

We have also measured the spectrum of the emitted light at a fixed $V_{SD}$ value, as a function of $V_G$, and found that in that case as well, the spectrum depends strongly on the device operation point. Figures 6a-d show the spectrum of the light emitted by a device realized on a 2L-WS$_2$/4L-InSe interface, as a function of gate voltage, for two different values of source-drain bias ($V_{SD} = +2.4$ V a-b and $V_{SD} = +3.12$ V c-d). Changing the gate voltage at fixed $V_{SD}$ allows switching the spectrum of the light between two transitions visible in Figures 6b,c. In particular, at large positive gate voltage ($V_G > +0.4$ V in Fig. 6a and $V_G > +0.8$ V in Figure 6c), the spectrum of the emitted light is dominated by the interlayer transition at 1.2 eV between the bottom of the InSe conduction band and the top of the TMD valence band. At low gate voltage ($V_G < +0.2$ V in Figure 6a and $V_G < +0.6$ V in Figure 6c), instead, light is emitted by another transition (possibly by multiple transitions, as suggested by the broad linewidth) at higher energy (approximately 1.4 eV), which appears to blue shift upon increasing $V_G$. Unexpectedly, the two regimes are separated by an interval of gate voltages in which the power of emitted light vanishes (or is below the sensitivity of our detector). Finding that the gate allows switching the spectrum between two different emission lines is extremely interesting, as it may provide new functionality to these LEFETs devices.

Our observation that the EL spectrum does depend on the device operation point proves that LEFETs based on vdW interfaces are indeed electrically tunable light sources. Understanding in detail how the EL spectrum depends on the LEFET operation point is however complex, both because different processes likely play a role, and because screening due to charges accumulated in the transistor channel can strongly (and non-linearly) affect the potential difference between the two layers forming the interface. At sufficiently large $V_{SD}$, we expect that electrons are injected not only in the conduction band of InSe but also in that of the TMD, so that light can be emitted also from intralayer transitions within the TMD. This may account for the peak centered around 1.6 eV in 2L-WS$_2$/4L-InSe, which corresponds well to one of the 2L-WS$_2$ PL peaks. An intralayer transition in the semiconducting TMD is likely also responsible for part of the broad peak around 1.5 eV in the MoS$_2$-based interfaces (the energy matches one of the peaks observed in PL of 2L-MoS$_2$). The less pronounced peaks near 1.4 eV (at comparable but different energies in the 2L-WS$_2$ and the 2L-MoS$_2$ devices; see Figure 5b and 5e) occur at an energy that changes slightly upon changing $V_{SD}$. That is why we tentatively attribute the peak to an interlayer transition –possibly an electron in a higher energy InSe sub-band, recombining with a hole in the TMD– whose precise energy is affected by the electrostatic potential difference between the layers. This attribution is also consistent with data taken at fixed $V_{SD}$ upon varying $V_G$ (see Figure 6), in which the transition energy is seen to blue shift upon changing the gate voltage.
More work is clearly needed to understand in detail the electroluminescence spectrum of Γ-Γ interfaces at large biases, as well as its evolution with both source-drain and gate biases. We note, however, that the spectrum of light emitted by LEFETs based on monolayer TMDs remains unchanged even under driving the device with very large source-drain biases, as discussed in Supplementary Section S4 and shown in Figure S4 in there. The data, therefore, appear to substantiate our initial idea—namely that LEFETs based on vdW interfaces offer more functionalities than similar devices realized from individual monolayers—irrespective of the precise microscopic origin of the emitted light (i.e., of the specific transitions involved in the light emission process).

Conclusions and outlook

The results presented above demonstrate the operation of vdW interface LEFETs and show that these transistors do allow the realization of light sources with a bias tunable spectrum. Finding that devices realized with multiple semiconducting TMDs and with InSe layers of different thickness lead to a qualitatively similar evolution of the spectrum of the emitted light with bias indicates that the operation mechanism is robust and of general validity. This robustness is important because—whereas only a few examples of electroluminescent devices with an electrically controllable spectrum have been reported in the past46–49—a large variety of 2D semiconductors exists that can be employed to realize light-emitting Γ-Γ interfaces.

It is clear that at this stage light-emitting transistors based on Γ-Γ vdW interfaces remain proof-of-principle devices and that considerable research is needed to characterize them and optimize their operation. We anticipate, for instance, that ionic liquid gating can be replaced by conventional solid-state gates using nm-thick h-BN dielectrics to separate the gate electrode from the vdW heterostructure. Indeed, h-BN layers that are a few nanometers thick exhibit breakdown field values approaching 1 V/μm,50 which are likely sufficient to achieve ambipolar transport and to operate the devices with $V_{SD}$ larger than $V_G$. Similarly, the details of the light emission processes (related to the optical selection rules for interlayer and intralayer transitions23,51) will have to be understood, and the light outcoupling investigated and improved. It is nevertheless worth emphasizing that Γ-Γ interfaces used within a light-emitting transistor configuration represent the first platform that becomes available for the realization of electroluminescent devices with bias tunable spectrum, which satisfies many key requirements essential to the development of a successful technology. These include room-temperature operation and insensitivity of the devices to details of their assembly process, which ensures their robust operation. It is for these reasons that exploring the development of light-emitting transistors based on Γ-Γ interfaces appears to be extremely promising for future device applications.

Acknowledgements

We gratefully acknowledge Alexandre Ferreira for continuous and precious technical support. AFM gratefully acknowledges financial support from the Swiss National Science Foundation (Division II) and from the EU Graphene Flagship project. VF acknowledges support from the European Quantum Flagship project 2D-SIPC and EPSRC grant EP/S030719/1. L.B. acknowledges support from US NSF-DMR 1807969 (synthesis, physical characterization, and heterostructure fabrication) and the Office Naval Research DURIP Grant 11997003 (stacking under inert conditions). L.B. acknowledges the use of of the facilities at the Platform for the Accelerated Realization, Analysis, and Discovery of Interface Materials (PARADIM) which is supported by the US-NSF under the Cooperative Agreement No. DMR-2039380. DS acknowledges support from the U.S. Department of Energy (DE-FG02-07ER46451) for photoluminescence measurements of InSe. DS and CNL acknowledge the support by NSF/ECCS award 2128945. The National High Magnetic Field Laboratory acknowledges support from the US-NSF Cooperative agreement Grant number DMR-1644779 and the state of Florida. K.W. and T.T. acknowledge support from the Elemental Strategy Initiative conducted by the MEXT, Japan (Grant Number JP-MX0112101001) and JSPS KAKENHI (Grant Numbers 19H05790, 20H00354 and 21H05233).

Author contributions

H.H., D.M., D.D., and M.P. fabricated the devices with the help of I.G.L.; H.H., and N.U. measured the devices and analyzed the data. S.M, W.Z. and L.B. grew the InSe crystals. Z.L., D.S., C.N.L., and D.S. performed photoluminescence measurements on hBN encapsulated InSe multilayer, to characterize their quality. V. F. identified the high quality of the InSe crystals and suggested their use the realization of different devices. K.W. and T.T. provided the hBN crystals. H.H., I.G.L., N.U., and A.F.M analyzed and discussed the data. A.F.M. wrote the manuscript with input from N.U., H.H., and I.G.L. All the authors read and commented on the manuscript. N.U. and A.F.M. supervised the research.
Competing financial interests

The authors declare no competing financial interests.

Data availability

The data supporting the findings of this study are available free of charges from the Yareta repository of the University of Geneva (https://yareta.unige.ch/).

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