Supporting Information

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2+\delta-Dimensional Materials via Atomistic Z-Welding

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\textbf{Fig. S1} (a) Schematic diagram of sonochemical exfoliation processes for graphene sheets in IPA solution. (b) Camera image of a sonochemical hybridized sample of Gr-BN with different wt. % (0.5 to 1) from left to right.
Fig. S2  Atomic profile for hybridized Gr-BN samples obtained from HRTEM, measured along with atomic arrays of atoms (horizontal) and across the atomic arrays of atoms (vertical) for (a) 1 wt. % and (b) 0.5 wt. %.

Section 1: XRD details of RGO-RBNO, MoS$_2$-WS$_2$, and MoS$_2$-RBNO system

A hybridized sample of RGO-RBNO was studied using the X-ray diffraction technique (XRD) to have a glimpse of inter-layer coupling and find out new hybridized peaks due to C-B and C-N bond formation. When XRD measurement was performed on the GBNH sample, we observed 2θ° peaks at 26.5°, 41.4°, 43.6°, 54.9°, 59.4°, 71.2°, and 75.8°. Similarly, GBNS exhibited XRD peaks at 26.7°, 41.4°, 43.9°, 56.1°, 59.5°, 71.3°, and 75.9°. When microwave power was used as an exotic condition (temperature and electric field) for hybridizing and obtaining product was diagnosed with XRD measurement, it exhibited peaks at 26.60°, 41.4°, 43.7°, and 55.0°, respectively 59.7°, 71.3°, and 75.7°. XRD peak at ~26.0° of B.N. corresponds to {002} facet of hexagonal closed pack structure shifted from their original position 0.5° for GBNH, 0.7° for GBNS, and 0.6° for GBNM sample. We observed a change in inter-layer d-spacing from ~3.42 Å to 3.3 Å for the GBNM sample, which was ~2.1% less than their original d-spacing. XRD is a bulk characterization technique that gives information at a macro level; any minute changes in the micro-level will not be detected by XRD. However, the Shift in micron level is moderately significant; we expect more changes would have occurred at the local atomic level, and M.D. simulation results support the same; the local distance was changed from 3.3 Å to 2.7 Å at the
Hybridized location. We also have found a distinct XRD peak at 54.9° due to diffraction from localized B₄C Like- crystal structure formation upon hybridization between constitute layers³, 23-24.

A similar hybridization technique was employed for semiconductor MoS₂ and insulator RBNO sheets, which have contrasts in their electronic and optical behaviors. The hybridized sample is expected to exhibit bandgap tuning, charge carrier density enhancement, and the emergence of new bond formation (Mo-N and Mo-B) upon hybridization. When XRD was used to examine the heating method's hybridized sample, various peaks were observed at 14.2°, 28.7°, 32.5°, 33.4°, 35.8°, 39.4°, and 43.8°, respectively 49.6°, 55.0°, and 59.7°. A similar measurement was performed for the solvothermally hybridized sample; it exhibited peaks at 14.3°, 26.6°, 28.9°, 32.6°, 33.4°, 35.8°, and 39.4°, respectively 44.0°, 49.7°, 55.0°, and 56.0°. Whereas, microwave treated hybrid sample demonstrated peaks at 14.4°, 26.8°, 29.0°, 32.7°, 33.5°, 35.9°, 39.5°, 44.1°, 49.8°, 55.1°, 56.1°, and 60.1°. Interestingly peak at 13.9° of {002} facet of the hexagonal MoS₂ phase shifted upon hybridization. The observed shift in the peak was 2.1 % for heating and 2.8 % for solvothermal, and 3.5 % for microwave-hybridized samples. The maximum change observed in inter-layer d-spacing was 6.14Å for the microwave-hybridized sample compared to 6.3 Å for pure 2H MoS₂. We also have witnessed distinct diffraction peaks at ~34.5° and 60.1° due to the Mo₂B like-structure. Similarly, peaks at ~39.4° and 44.0° signified the diffraction peaks due to MoN like-structure. The presence of various bond (Mo-N and Mo-B) in XRD suggest strong hybridization between MoS₂ and B.N. layers under microwave and solvothermal processing condition²,5-7.
Fig. S3 XRD pattern (a) small scale scan for GBNH, GBNS, and GBNM samples and (b) large range scan for GBNH, GBNS, and GBNM samples. (c-d) Small scale and extensive scan range for MBNH, MBNS, and MBNM samples. (e-f) small scale and extensive scan range for MWN, MWS, and MWM samples.

MoS$_2$ and WS$_2$ being semiconductors, with bandgap (1.4 to 1.9 eV), both exhibit direct band gaps only when synthesized in monolayer. Synthesizing the monolayer of MoS$_2$ and WS$_2$ using scotch tape and sonication is a cumbersome process, whereas CVD is expensive and non-scalable. Overcoming such hurdles, we combine these two-dimensional materials to tune their excitonic behaviors by hybridizations (heating, solvothermal, and microwave). XRD measurement was performed for hybridized (MoS$_2$-WS$_2$) sample by MWH exhibited peaks at 14.4º, 28.9º, 31.2º, 32.7º, 33.5º, 35.8º, 39.5º, 43.9º, 49.8º, 56.0º, 58.4º, 59.8º, 62.7º, 70.2º, 73.0º, and 80.1º. Likewise, for the MWS sample, it exhibited peaks at 14.3º, 26.6º, 29.0º, 31.4º, 32.6º, 33.5º, 35.8º, 39.5º, 41.6º, 44.1º, 49.8º, 50.07º, 56.0º, 58.3º, 60.1º, 62.7º, 70.1º, 72.7º and 80.2º. To distinguish microwave processing, which involves an electric field, from solvothermal hybridization (pressure and temperature), we carried out XRD measurement in the microwave process MWM sample. We observed peaks at 14.1º, 28.8º, 31.4º, 32.6º, 33.5º, 35.7º, 39.4º, 43.9º, 49.7º, 56.0º, 58.2º, 59.8º, 62.7º, 70.0º, 72.8º, and 80.2º. Characteristic peak
at 14.32° of (002) facet of WS$_2$(2H phase) hexagonal phase-shifted to 14.1° in the microwave-hybridized sample is also distinct peaks were observed in X-ray diffraction at 29.0°, 44.0°, and 60.1° due to hybridization between MoS$_2$ and WS$_2$. Inter-layer d-spacing was reduced to 6.2 Å in contrast to 6.3 Å for pure WS$_2$ slight reduction in inter-layer d-spacing was observed because of the global stacking effect. Still, at the local level, it is expected to reduce even higher.

**Section 2 Raman and FTIR measurement**

The 2D atomic layer constitutes atoms bound with strong covalent bonds (in-plane), whereas the out-of-plane direction exhibits van der Waals's nature. When hybridized between RBNO, RGO, MoS$_2$, and WS$_2$ is exhibited various bond formations. These bonds can be catalyzed by external perturbation by optical and electrical energy and will suffice for atomic rotational and vibrational transitions. Thus, various characteristics of vibration bond energy will be observed when diagnosed with Raman and Fourier transfer infrared (FTIR) spectroscopies. When FTIR measurement was done for GBNM sample, it exhibited vibration mode at 790, 926.9, 1088.7, 1180.9, 1238.8, 1320.1, 1483.5, 1727.6, 2313.5, 2828.0 and 2973.2 cm$^{-1}$. Similarly, the GBNS sample exhibited vibration modes at 752, 875, 927.7, 1096.2, 1320.9, 1494.5, 1720.0, 2539 and 2985.3 cm$^{-1}$. Likewise, for GBNH sample, varied rotational peaks were observed at 725.5, 903.7, 1041.2, 1269.2, 1330.4, 1447.08, 1542.88, 1725.19 and 2328.7 cm$^{-1}$. Typically, ~750 to the 760 cm$^{-1}$ represents an out-of-plane bending mode of B-N-B, and 1320 cm$^{-1}$ is due to the B-N stretching mode. After hybridization, a new vibration mode at 1727.6 cm$^{-1}$ (C-N) was observed. Similarly, a bond at (~ 1212-1270) cm$^{-1}$ was found due to C=N vibration$^{13-14, 16-17}$. A bond at 2928 cm$^{-1}$ was due to symmetric stretching mode, and 2973 to 2985 cm$^{-1}$ was due to asymmetric stretching mode of CH$_3$ after reducing functional group COOH to CH$_3$.
Raman spectroscopic measurement was performed in the GBNH sample. After deconvolution, it exhibited Raman peaks at 1235 cm\(^{-1}\) due to hybridized (C-N) bond formation, 1322 cm\(^{-1}\) (D band), 1365 cm\(^{-1}\) due to B-N vibration mode, 1574 cm\(^{-1}\) (G band) 1599 cm\(^{-1}\) (D’ band), 2552 cm\(^{-1}\) (D+D’), 2642 cm\(^{-1}\) (2D band) and 2669 cm\(^{-1}\). Likewise, GBNS sample exhibited Raman vibration modes at 1266 cm\(^{-1}\), 1328 cm\(^{-1}\) (D band) and possibly due to B-C vibration mode\(^8\).-\(^10\), 1363 cm\(^{-1}\) (B-N vibration mode), 1547 cm\(^{-1}\) (G band), 1607 cm\(^{-1}\) (D’ band), 2519 cm\(^{-1}\) (D+D’), 2586 cm\(^{-1}\) (2D band) and 2702 cm\(^{-1}\). Similarly, for the GBNM sample, in addition to pressure and temperature, a high electric field (~ 10\(^6\) V/cm) led to the strong inter-layer coupling between (RGO-RBNO) system when it was diagnosed with Raman measurement. We observed different peaks at 1319 cm\(^{-1}\) due to (C-N)\(^{11}\) bond formation 1350 cm\(^{-1}\) (B-N vibration), 1579 cm\(^{-1}\) (G band), 1603 cm\(^{-1}\) (D’), 2994 cm\(^{-1}\) (D+D’), and 2547 cm\(^{-1}\) (2D band)\(^{10,12}\). Hybrid materials are supposed to exhibit new vibration modes and splitting of Raman modes of an individual layer. The same has been observed after the deconvolution of 2D peaks. A similar effect was observed in D peak intensity, which increased due to hybridization. The effect electric field was observed during Raman measurement by varying the electric field value from 0 V to 13 V in the RGO-RBNO system. When the voltage value was zero, Raman peaks were observed at 1296 cm\(^{-1}\), 1351 cm\(^{-1}\) (B-N) vibration, 1586.7 cm\(^{-1}\) (G band), and 1614.9 cm\(^{-1}\) (D’ band). Similarly, when the voltage was changed to 5 V, it exhibited Raman peaks at 1314 cm\(^{-1}\) (C-N)\(^{11}\) vibration mode, 1365 cm\(^{-1}\) (B-N) vibration, 1411 cm\(^{-1}\), 1587.9 cm\(^{-1}\) (G band), and 1614.26 cm\(^{-1}\) (D’ band). Likewise, when voltage was
increased to 12 V, it displayed Raman modes at 1294.06 cm\(^{-1}\), 1340.49 cm\(^{-1}\) (B-C)\(^8\) vibration modes from B\(_4\)C like-structure, 1368 cm\(^{-1}\) (B-N) vibration mode, 1394.5 cm\(^{-1}\), 1578.8 cm\(^{-1}\) (G band), and 1616.9 cm\(^{-1}\) (D') band. When voltage was further increased to 13 V, it exhibited Raman modes at 1290 cm\(^{-1}\), 1319 cm\(^{-1}\) (C-N)\(^{11}\) vibration, 1364.7 cm\(^{-1}\) (B-N) vibration, 1576.3 cm\(^{-1}\) (G band), and 1615 cm\(^{-1}\) is due to (D') band. Inter-layer coupling and bond formation in the hybridized (RGO-RBNO) sample, influenced by an electric field and these vibration modes, were activated when we increased the voltage\(^{12,13}\). When Raman measurement was performed under an applied electric field, the G band (C-C bond stretching mode) in RGO due to sp\(^2\) carbon atoms was red-shifted as we increased the voltage from 0 V to 13V. Such red shifts in the G band signified electric field-induced doping in RGO-RBNO. Also, it suggests a strong modification of DOS with new states formation along with bandgap opening in the E-K energy band diagram. Also, linear E-K behavior changed parabolic nature at \(\Gamma\) point upon field doping.

**Fig. S5** (a) Raman spectra of BNH, GBNS, and GBNM hybridized samples (b-c) G and 2D peaks deconvolutions for GBNH, GBNS, and GBNM hybrids. (d) Electric field mediated Raman spectra for different voltages. (e) Raman spectra for MoS\(_2\) and B.N. hybridized (MBNN, MBNMI, MBNH) samples, (f) blue Shifts in B.N. peaks position in hybridized samples. (g) \(E_{2G}\) and \(A_{1G}\) peak positions in hybridized BNMH, BNMM, and BNMS samples. (h) \(\Delta \omega\) between \(E_{2G}\) and \(A_{1G}\) modes due to hybridization. The minimum was observed for the MBNMI sample. (i) Raman spectra for MoS\(_2\) and WS\(_2\) hybridized (MWH, MWS, and MWM) samples (j) \(A_{1G}\) vibrational position of MoS\(_2\) (k) \(\Delta \omega\) difference between \(E_{2G}\) and \(A_{1G}\) modes for
MoS$_2$ due to hybridization. (l) Intensity ratio for $A_{1g}/E_{2g}$ peaks for MoS$_2$ samples when hybridized with WS$_2$.

Similarly, RBNO was hybridized with MoS$_2$ with an adequate supply of energies (temperature, pressure, and electric field energy). These layers (RBNO and MoS2) are expected to come close to each other as an effect of Mo-N, Mo-B, and N-S bond formation is desired. When hybridized sample BNMH was diagnosed with FTIR measurement, we observed different peaks at 759, 1160, 961.36, 1322.9, 2355.1, and 2330 cm$^{-1}$. Similarly, for BNMS sample, it exhibited FTIR peaks at 768, 1375.5, 1166, 962.2, 2338, 2545.3, 2527.4, and 2603 cm$^{-1}$. Likewise, the BNMM system displayed FTIR peaks at 761, 952.4, 1169.8, 1329.2, and 2352 cm$^{-1}$. We have also performed the FTIR measurements for 2D B.N. exfoliated sheets to check shifts in vibration modes compared to hybridized samples. 2D B.N. sheet exhibited FTIR peaks at 758 cm$^{-1}$ due to B-N-B out-of-plane bending mode and 946, 1327 cm$^{-1}$ due to B-N stretching mode. The out-of-plane vibration mode of B-N blue-shifted 10 cm$^{-1}$ in BNMM hybridized sample, primarily due to interaction with neighboring MoS$_2$ sheets; and possibly out-of-plane bond formation. Their elastic behavior will change depending upon bonding nature (strong or weak); consequently, effective frequencies will shift from their original positions. Similar behavior was exhibited by the BNMS sample, where the B-N stretching mode got shifted by 8 cm$^{-1}$. Active sites available during the hybridization (B and N) are more likely to interact with MoS$_2$; due to the offset of S atoms from MoS$_2$ atomic sheets, the stretching vibration frequency of B-N will change.

**Fig. S6** FTIR spectra for B.N. and its hybrids with MoS$_2$(BNMH, BNMS, BNMM) (a) for a short scan range of 300 to 1800 cm$^{-1}$. (b) For full scan range for the hybridized (BNMH, BNMS, BNMM) samples.
When Raman measurement was performed on BNMH, it exhibited Raman peaks at 365 cm\(^{-1}\) due to hybridized (Mo-B) bond, 370 cm\(^{-1}\) (E\(_{2g}\) mode), 400 cm\(^{-1}\) (A\(_{1g}\) mode), and 449 cm\(^{-1}\) (2LA mode) modes of MoS\(_2\) and 1357 cm\(^{-1}\) are due to B-N. It is noteworthy that a systematic blue Shift was observed in B-N (1350 cm\(^{-1}\)) vibrational mode in Raman measurements. The shifts were 3.7 cm\(^{-1}\) for BNMH, 6.2 cm\(^{-1}\) for BNMS, and 7.1 cm\(^{-1}\) for BNMM samples. Similarly, difference \(\Delta \omega\) (E\(_{2g}\)-A\(_{1g}\)) modes of MoS\(_2\) due to hybridization were found at 30.2 cm\(^{-1}\) for BNMH, 26.8 cm\(^{-1}\) for BNMS, and 28.2 cm\(^{-1}\) for BNMM samples. Decreased \(\Delta \omega\) manifest hybridization and various bond formations (Mo-B and Mo-N). Also, we have found that phase transformation of MoS\(_2\) (2H to 1T) is more prominent in solvothermally hybridized samples compared to heat-mediated and microwave-based hybridized samples. We have also performed new Raman measurement in our hybrid samples and found that more prominent 1T phase Raman vibration modes \([36-37]\) at \(~219\) cm\(^{-1}\) and \(~315\) cm\(^{-1}\) for Solvothermally hybridized sample (BNMS) in contrast to heat mediated (BNMH) and microwave hybridized (BNMM) samples.

When MoS\(_2\) is hybridized with WS\(_2\), upon hybridization, from MoS\(_2\) Mo and S atom will interact with WS\(_2\) (both active sites W and S) and vice versa; as an effect, Mo\(_1\)xW\(_x\)S\(_2\) like- new crystal structure formation is expected. Raman vibration mode E\(_{2g}\) (in-plane vibration) of constituent atoms, whereas A\(_{1g}\) (out-of-plane) shift of S atom from existing (MoS\(_2\) and WS\(_2\)) atomic sheets are sensitive to measurements. When Raman measurement was performed in hybridized MWH (heating) sample, it exhibited a peak at 288, 319, 344, 373, 400, 415, 444, 516, 576, and 625 cm\(^{-1}\). Likewise, MWS (Solvothermal) sample exhibited Raman modes at 293, 338, 346, 375, 400, 416, 446, 577, 519 and 633 cm\(^{-1}\). Similarly, MoS\(_2\) was hybridized with WS\(_2\) by microwave (MWM), and the response will differ due to their different dielectric constants in the GHz frequency range. Also, a high electric field (~10\(^6\) V/cm) and localized heating (~2000 \(^\circ\)C). Although hybridization was performed in the liquid medium, it will push atomic sheets (MoS\(_2\) and WS\(_2\)) even closer to each other. As an outcome, the inter-layer coupling will enhance. When Raman measurement was performed it revealed various vibration modes at 286, 322, 345, 373.5,400,414,444,516,577and622cm\(^{-1}\). Generally, exfoliated MoS\(_2\) sheets exhibits Raman modes at370cm\(^{-1}\) (E\(_{2g}\)), 400cm\(^{-1}\)(A\(_{1g}\)), 452cm\(^{-1}\) (2LA), 517cm\(^{-1}\) (E\(_{2g}+2LA\)), 577cm\(^{-1}\) (E\(_{2g}'+LA\)) and 622 cm\(^{-1}\) (A\(_{1g}+LA\)). Similarly, exfoliated WS\(_2\) sheets exhibit Raman modes at 175 cm\(^{-1}\) (LA),265cm\(^{-1}\) (2LA-3E\(_{2g}\)),230cm\(^{-1}\) (A\(_{1g}-LA\)),296cm\(^{-1}\) (2LA-2E\(_{2g}\)),352cm\(^{-1}\) (2LA),355(E\(_{2g}'+\)), 418 cm\(^{-1}\) (A\(_{1g}\)), 581 cm\(^{-1}\) (A\(_{1g}+LA\)) and 702 cm\(^{-1}\) (4LA). In general, Raman E\(_{2g}\) (in-plane vibration) and A\(_{1g}\) (out-of-plane vibration) modes are considered the most important Raman signature in exfoliated MoS\(_2\) and WS\(_2\) sheets and therefore were diagnosed minutely in hybridized sheets, and their effect on hybridization was studied\([4,18-23]\). The difference (E\(_{2g}\)-A\(_{1g}\)) in Raman modes, i.e., \(\Delta \omega\) for MoS\(_2\) was 27.24 cm\(^{-1}\) for MWH and 25.8 cm\(^{-1}\) for MWS hybridized sample. Likewise, for WS\(_2\) difference (E\(_{2g}\)-A\(_{1g}\)) \(\Delta \omega\) was found to be 70.54 cm\(^{-1}\) for MWH and 68.9 cm\(^{-1}\) for the MWM sample. Such a significant Shift hint at the hybridization and bond formation between the MoS\(_2\) and WS\(_2\) system; as an effect, its intensity ratio (A\(_{1g}/E_{2g}\)) was changed.
Section 3: TEM analysis of RBNO-MoS$_2$ and MoS$_2$-WS$_2$ system

Fig. S7 Camera image of (a) synthesized BNO and G.O. sample on a large scale. (b) Mixed G.O. and BNO for hybridization with different wt. % (20, 50, and 80) in DMF. (c) Hybridized sample after processing by the heating method with wt. % (50:50) of RGO-RBNO. (d) Microwave- hybridized RGO on RBNO with different wt. % from left to right (20 %, 50 % and 80 %).

Fig. S8 (a-c) TEM elemental mapping of GBNH, GBNS, and GBNM samples. (d-f) Atomic line profiles were obtained from HRTEM and the atomic arrays (horizontal) and across the atomic arrays (vertical) for hybridized GBNH, GBNS, and GBNM samples.
Like the RGO-RBNO hybrid system, it is very intriguing to check hybridization and its corresponding change in the atomic arrangements of hybrid RBNO-MoS$_2$ systems. Therefore, we hybridized RBNO with MoS$_2$ by heating, solvothermal, and microwave (see camera image in Fig. (S9)). When the BNMH sample was diagnosed with TEM (see Fig. S10 (a)), we observed a highly transparent sheet corresponding to the MoS$_2$ sheet covered with oval-shaped 2D sheets of B.N. sheets. The overlapped area was estimated at $\sim$ 200 nm, which was further validated by elemental mapping, which gave clear proof, i.e., the presence of both MoS$_2$ and RBNO sheets. When the overlapped area was minutely diagnosed with HRTEM moirés pattern was observed, which signifies the hybridization between MoS$_2$ and RBNO (due to inter-layer coupling). When FFT was acquired in the overlapped area of HRTEM, it exhibited mixed-fold symmetry (6-fold with 2-fold) (see inset Fig. S10 (b)). Such variation in symmetry suggests the influence of foreign atomic sheets (MoS$_2$) over RBNO sheets. The average nuclear distance was measured in the HRTEM image across the atomic line and found to be $\sim$2.5 Å, whereas, along the atomic line, it was $\sim$2.1 Å. Signify the highly strained system due to strong inter-layer coupling. In contrast, the equilibrium distance for B.N. is $\sim$1.54Å, and for MoS$_2$, it is $\sim$3.1 Å. Similarly, the BNMS sample was examined by TEM imaging highly transparent, and folded MoS$_2$ sheets were observed. This is characteristic of monolayer MoS$_2$ along with oval-shaped B.N. sheets. The overlap area was estimated to be $\sim$150 nm, confirmed by elemental mapping. When the overlying area was examined by HRTEM, we could resolve the individual atoms; the atomic arrangements also suggested a moirés pattern, possibly due to overlapping between distinct atomic sheets with some rotational angle. FFT pattern in an overlap area exhibited six-fold symmetry, possibly due to A.A. stacking. Measuring the atomic distance in the obtained moirés pattern will be interesting, as it will have different distances than their parent atomic sheets; hence we measured average atomic distances. Along the atomic line, it was $\sim$3.45Å; across the atomic line, it was 3.0Å. It signified that MoS$_2$ atomic distance had
It has been reduced ~ 3% significantly from its equilibrium distance of 3.15 Å; due to the presence of B.N. sheets in the vicinity of MoS$_2$ sheets.

Moreover, the MBNMI hybrid sample was hybridized in the liquid medium, where the localized temperature was moderately high with an extremely high electric field. Such dynamic conditions excelled RBNO and MoS$_2$ sheets close enough where bond formation occurred. When the hybridized sample was examined by TEM measurement, it exhibited transparent sheets with sharp-edged MoS$_2$ sheets and oval-shaped B.N. sheets extending up to more than 200 nm in lateral dimension, with an overlap area of ~100 nm. The presence of MoS$_2$ and RBNO in the overlap area was further confirmed by elemental mapping.
The overlap area was revisited by HRTEM measurement; atoms were linearly arranged in some places (plausibly moirés pattern), whereas it exhibited hexagonal patterns in some places. When the FFT pattern was observed, it showed 6-fold symmetry. Localized change in inter-atomic distance will be exciting because it will alter its electronic and optical properties. Therefore, we carried out atomic distance measurements obtained from the HRTEM image. Along the atomic line, it was ~3.2 Å, and across the atomic arrangement, it exhibited ~3.0 Å.

Fig. S11 Elemental mapping of RBNO-MoS$_2$ hybridized samples and their atomic line profiles obtained from the HRTEM images along with the atomic array (lateral) and across the atomic array (vertical). (a, d) for BNMH (b, e) for BNMS (c, f) for BNMM.

MoS$_2$ and WS$_2$ are of great interest due to their potential applications in optoelectronics, photocatalysis, sensors, and photovoltaics. Hybridizing such 2D materials will unveil various applications starting from energy storage, excitonics, transistors, and many more. Conventional hybridization techniques, e.g., CVD grow sheets, wet chemical transfer, and scotch tape transfer methods, have some limitations for device applications. The diffused interfaces during growth, residual contamination (water molecules and glue), and scalability are major concerns. We have tried to address the existing problem by exfoliating a few layers of MoS$_2$ and WS$_2$ in large quantities and hybridizing by heating, solvothermal, and microwave (see camera image of hybridized sample Fig. S12). The resulting hybridized sample was diagnosed by TEM imaging to know the effect of hybridization at the atomic scale. When the hybridized MWH sample was
Analyzed with TEM, observed highly transparent sheets with lateral dimensions of more than ~150 nm correspond to MoS$_2$ sheets, covering WS$_2$ sheets of ~30 nm (see Fig. S13 (a)). The presence of both atomic sheets in the overlap area was confirmed by elemental mapping (see Fig.S12 (d)). When overlapping areas 1& 2 were minutely diagnosed with HRTEM imaging, we noticed a hexagonal pattern in a few places along with a moirés pattern (characteristic of inter-layer coupling) that suggests interaction leading to bond formation amongst atomic sheets. The hybridized area was further studied by measuring the interatomic distance and found across the atomic line ~3.15Å and along the atomic line ~3.0 Å. Similarly, MWS hybridized sample was diagnosed by TEM imaging; we observed highly transparent folded WS$_2$ sheets of ~ 200 nm wrapping the MoS$_2$ sheet, which were tiny sheets of ~ 50 nm (See Fig.S13 (b)). Elemental mapping confirmed the coexistence of MoS$_2$ and WS$_2$ sheets. When the overlying areas 1&2 were minutely examined by HRTEM measurement, they exhibited the atomic arrangement in a hexagonal manner. The FFT pattern further validated this; it showed (6-fold symmetry). Atomic distances obtained from HRTEM along the atomic line were ~3.15Å, whereas, across the atomic line, it was ~3.0Å. When MWM hybridized sample was examined by TEM measurement, multilayer highly transparent sheets were observed lengthened up to ~250 nm, covering the WS$_2$ atomic sheets ~30 nm (see Fig.S13 (c)). Elemental mapping revealed the presence of both MoS$_2$ and WS$_2$ sheets. When such overlapped areas 1&2 were diagnosed with HRTEM, it exhibited a moirés pattern over the entire area; it signifies the strong inter-layer coupling as an effect of high electric field and supercritical heating under liquid medium. FFT pattern was performed over the underlying area; it exhibited six-fold symmetry, which suggests possible A.A. stacking. Inter-atomic distances were measured over the moirés pattern vertically and horizontally from the HRTEM image; it turned out that the vertical distance was ~3.15 Å and was larger than the horizontal distance of ~ 3.0Å.
Fig. S12 (a-b) Camera image of large-scale synthesis of MoS$_2$ and WS$_2$ 2D sheets and its hybrid, (c) camera image of processed MWH hybrid sample (d-f) elemental mapping of hybridized (MWH, MWS, and MWM1) samples.
Fig S13 (a) TEM image of MWH, overlapped areas are marked as 1 and 2 and corresponding HRTEM inset their FFT. (b) TEM image of MWS, overlapped areas are marked as 1 and 2 and corresponding HRTEM inset their FFT (c) TEM image of MWM, overlapped areas are marked as 1 and 2 and corresponding HRTEM inset their FFT.
Section 4: XPS details of RGO-RBNO RBNO-MOS₂ and MoS₂-WS₂ hybridized sample

Effect of hybridization on hybridized samples (GBNH, GBNH, and GBNM) attained by various processing conditions (heating, solvothermal, and microwave) and their effect on interlayer distance and the interlayer coupling in-depth. Due to dynamic processing conditions is very important to diagnose with XPS for better understanding. XPS measurements were performed on hybridized GBNH sample (Survey shown in Fig.S15 (a)) and a short-range scan for B1s, C1s, and N1s spectra (see Fig.S15(b)-(c)). Upon deconvolution of B1s, spectra’s stretches at 189.6 eV (B-C), and 190.9 eV (B-N) bonds were obtained. Similarly, after deconvolution of C1s spectrum, we observed stretches at 282.8 eV (B-C) B₄C like-structure, 283.4 eV (B-C) BC₃ like-structure, 284.2 eV (C-C) bond, 285.5 eV (C-N) bond, and 288.0 eV corresponding to C-O bond. Likewise, the N1s spectrum after deconvolution exhibited stretches at 397.1 eV corresponding to the B-N bond, 98.4 eV C-N pyridinic like a bond, and 399.3 eV (C=N) pyrrolic like a bond. Inter-layer coupling was expected to get even stronger when additional dynamic condition pressure was involved in hybridization. Therefore, we carried out an XPS measurement of the GBNS sample (survey shown in Fig.S15 (d)) and a short-range scan for B1s, C1s, and N1s spectra (See Fig. S15 (e)-(f)). Upon deconvolution of B1s spectra, stretches at 189.7 eV (B-C) due to BC₃ like-structure and 190.6 eV due to sp² bonded (B-N) were obtained. Likewise, after deconvolution of the C1s spectrum, we observed stretches at 283.5 eV (B-C) BC₃ like-structure, 284.4 eV due to sp² hybridized C-C bond, 285.1 eV is because of C-N and 286.60 eV Corresponding to C-O bond. Similarly, the N1s spectrum after deconvolution exhibited stretches at 397.2 eV corresponding to the B-N bond, 398.0 eV C-N due to the pyridinic bond, and 405.5 eV because of the N-O bond. When microwave was employed for hybridization, dielectric heating due to oscillating electric field and high electric field will catalyze the reaction, and inter-layer distance will be further reduce. When XPS measurement was performed on GBNM hybrid sample (see survey in Fig. S15 (g)), and short-range scans for B1s, C1s, and N1s spectra (see Fig. S15 (h-i)). Upon deconvolution of B1s spectra, stretches at 189.7 eV (B-C) bond and 190.7 eV (B-N). Likewise, the N1s
spectrum after deconvolution exhibited stretches at 397.2 eV corresponding to the B-N bond, 398.0 eV (C-N) due to pyridinic-like bond, and at 405 eV (N-O) like a bond. Similarly, after deconvolution of C1s spectrum exhibited different stretches at 283.0 eV (B-C) BC$_3$ like-structure 284.0 eV due to sp$^2$ bonded C-C, 285.1 eV due to pyridinic like structure (C-N) and at 287.6 eV corresponding to N-C=O bond were obtained. Various bond % due to different techniques of hybridization were estimated and it was found that the B-C bond was 31 % for GBNH, 37.5 % for GBNS, and 5 % for GBNM hybridized sample. Likewise, the same calculation was performed for the C-N bond, and it was found to be 24 % for GBNH, 27.1% for GBNS, and 50.2% for the GBNM sample. We also observed that B-N bonding % decreased from 27 % for GBNH to 23 % for GBNM hybridized sample because B and N participated in bond formation with C atoms$^{11,14,15-16}$. 
**Fig. S15** XPS spectrum for RGO-RBNO hybridized samples full range scan (a) for GBNH, (d) for GBNS, and (g) for GBNM hybrid samples. (b-c) short scan range for B 1s and N 1s spectra for GBNH. (e-f) short-range scans (180 to 196 eV) for B 1s and N 1s (390 to 410 eV) spectra for the GBNS sample. (h-i) short-range scans (180 to 196 eV) for B 1s and N 1s (390 to 410 eV) spectra for the GBNM sample. (j) B-N bonding % estimated for GBNH, GBNS, and GBNM obtained from B 1s, and N 1s spectra, and (k) B-C bonding % calculated for 1 GBNH, 2 GBNS, and 3 GBNM obtained from B 1s and C 1s spectrum and (l) C-N bonding % GBNH, GBNS, and GBNM derived from C 1s and N 1s spectra. Similarly, the hybridized BNMH sample was diagnosed by XPS measurements (survey showed in Fig.S16 (k)) and short-range scan for B 1s, Mo 3d, N 1s, and S 2p spectra (See Fig. S16 (l)-(o)). Upon deconvolution of the B 1s spectrum, stretches at 189.2 eV, 189.8 eV owing to Mo-B-like structure (Mo-B), and at 191.0 eV assigned to sp² structure of B-N (B-N) were obtained. Likewise, N 1s spectrum after deconvolution displayed stretches at 394.0 eV corresponding to Mo-N bond 397.1eV, 398.2 eV due to B-N, and 405.3 eV because of N-O bond. Likewise, Mo 3d spectrum exhibited stretches at 225.3 eV ascribed to Sulphur 2s, 227.5 eV, and 228.1 eV due to the Mo-B bond.
attributed to MoB₂ like-structure, 228.7 eV (3d_{5/2}) and 231.2 eV (3d_{3/2}) of metallic 1T phase of MoS₂, 231.7 eV (3d_{5/2}) ascribed to semiconductor 2H phase of MoS₂. Similarly, after the S 2P peak deconvolution, it exhibited stretches at 161.0 eV (2P_{3/2}) and 162.25 eV (2P_{1/2}) owing to the metallic 1T phase of MoS₂, 163.6 eV, and 167.8 eV is because of elemental Speak. Similarly, the BNMS hybrid was examined by XPS (survey shown in Fig. S16 (a)) and short-range scan for B1s, Mo 3d, N1s, and S2p spectrum (See Fig. (b)-(e)). Upon deconvolution of B1s spectra, stretches at 189.2 eV and 189.4 eV owing to Mo₃B-like structure (Mo-B) bond and 191.1 eV assigned to sp² like structure of B.N. (B-N) bond and 192.2 eV due to (N-O) bond. Likewise, N1s spectrum was deconvoluted to display stretches at 394.1 eV corresponding to (Mo-N) bond 397.0 eV, 398.2 eV due to (B-N), and 405.3 eV because of the N-O bond. Likewise, Mo 3d spectrum exhibited stretches at 225.4 eV assigned to sulfur 2s, 227.6 eV and 228.2 eV due to (Mo-B) bond attributed to MoB₂ like- structure, 228.7 eV (3d_{5/2}) and 231.2 eV (3d_{3/2}) ascribed of metallic 1T phase and at 231.8 eV (3d_{5/2}) which signifies 2H phase of MoS₂. Similarly, after the S 2P peak deconvolution, it exhibited stretches at 160.9 eV (2P_{3/2}) and 162.1 eV (2P_{1/2}) owing to the 1T phase of MoS₂, 163.6 eV, and 167.8 eV because of elemental Speak. Hybridized sheets obtained by microwave method (BNMM sample) were scrutinized by XPS (see survey in Fig. S16 (f)), and their short-range scan for B1s, Mo 3d, N1s, and S2p spectra (See Fig. (g-j)). Upon deconvolution of B1s spectrum, stretches at 189.9 eV due to Mo₃B-like structure Mo-B bond and 191.1 eV ascribed to sp² like structure of B.N. (B-N) bond. Likewise, for the N1s spectrum after deconvolution, it exhibited stretches at 394.3 eV corresponding to Mo-N bond, 397.4 eV, 398.7 eV due to B-N, and 406.6 eV because of the N-O bond. Likewise, Mo 3d spectra displayed stretches at 225.7 eV ascribed to sulfur 2s, 228.2 eV (Mo-B) bond attributed to MoB₂ like- structure, 228.7 eV (3d_{5/2}) and 231.2 eV (3d_{3/2}) signifies 1T phase of MoS₂, 232.7 eV (3d_{5/2}) ascribed 2H phase of MoS₂. Similarly, after deconvolution of the S 2P peak, it exhibited stretches at 161.2 eV (2P_{3/2}) and 162.8 eV (2P_{1/2}) owing to the metallic 1T phase of MoS₂, at 162.4 eV (2P_{3/2}) due to 2H phase of MoS₂ and at 167.8 eV is because of elemental S peak.
**Fig. S16** XPS spectrum for (MoS$_2$-RBNO) hybridized samples full range scan (0-1400 eV) for (a) BNMS, (f) BNMM, and (k) BNMH (b-e) short-range scans for B1s, N 1s, Mo 3d, and S 2p for BNMS sample (g-j) short-range scans for B1s, N 1s, Mo 3d and S 2p for BNMM sample and (l-o) short-range scan for B1s, N 1s, Mo 3d and S 2p for BNMH sample.

**Fig. S17** (a-d) DFT band structure calculations for Gr-BN hybridized samples for A.A. stacking sequence for two distances 4.5 and 3 Å.
Fig. S18 Density of state for Gr-BN hybridized samples for A.A. stacking for two distances (a) 3.0 Å (b) 2.7 Å and Gr-BN AB stacking sequence with inter-layer distances (c) 3 Å and (d) 2.7 Å.
Fig. S19 (a) Tauc plot for GBNH, GBNS, GBNM, (b) Logarithmic resistivity vs. (1000/temperature) graph for RGO-RBNO hybrid (GBNH, GBNS, and GBNM) samples.
Fig. S20 Resistance Vs. (1000/temperature) graph for MoS$_2$-RBNO hybridized (BNMH, BNMS, and BNMM) samples.
Fig. S21 Hall coefficient measurements for RGO-RBNO hybridized samples (a) GBNS (b) GBNM (c) for GBNH (d) Carrier density graph for GBNH (N), GBNS (S), and GBNM (Mi).
Fig. S22 Charge density difference profiles for conduction band and valance band when the inter-layer distance was 2.7Å for Gr-BN system at different dispersion (K, M, and Γ ) points (a-c) Side view and (d-f) top view.
Fig. S23 DFT band structure calculation for BN-MoS\textsubscript{2} hybrid when the inter-layer distance was reduced from 3.3 Å to 2.7 Å, exhibited band gap was increased from 2.7 eV to 2.9 eV.

Section 5: Bond formation energies of various bonds and suitability of the techniques

In our hybridization techniques, such as the solvothermal technique, pressure is the main candidate that decides the chemical bond formation between the different atomic sheets. The pressure built up inside the solvothermal reactor depends upon the choice of solvents and temperature of the reactor. In our case, we've chosen DMF due to its reducing nature, and the reactor temperature was ~200°C. The experimentally available DMF pressure is ~18.806 kPa for 96 °C\textsuperscript{[26]}. Further pressure can be estimated by using Vangner type vapor pressure equation developed for DMF solvent

\[
\ln \left( \frac{p}{p_c} \right) = \frac{T_c}{T} \left( a_1 \tau + a_2 \tau^{1.25} + a_3 \tau^3 + a_4 \tau^7 \right) \quad \text{............... (1)}
\]

Where \( \tau = 1 - \frac{T}{T_c} \), \( a_1 = 36.48688 \), \( a_2 = -68.67427 \), \( a_3 = 62.2708 \), \( a_4 = -229.0057 \), \( T_c = 596.6 \text{ K} \) is the critical temperature, and \( p_c = 5.220 \text{ MPa} \) is the critical pressure\textsuperscript{[26]}

Using the above equation for \( T = 200 \text{°C} \) to the obtained vapor pressure of DMF within the reactor by substituting the constant values in the equation we obtained a pressure
is ~0.5 MPa. These conditions (pressure ~0.5 MPa and temperature ~200 °C) are sufficient for bond formation. As it was discovered in the case of graphite sp\(^2\) that gets converted to graphite-like Dimond (sp\(^2\)+sp\(^3\)) at elevated temperature and pressure. The P-T diagram for such conversion is shown in Fig.S24 \cite{27}.

Fig.S24. P-T phase diagram of carbon the light green color represents our experimentally obtained temperature and pressure zone, where the chemical bond formation occurs.

Similarly, the microwave-based processing method for hybridizing different atomic sheets depend upon microwave power. The microwave power absorption relies upon the material's dielectric and magnetic properties (if the sample is magnetic in nature). The power absorbed by the material inside the microwave reactor in liquid medium is given by

\[
P = 2\pi f (\varepsilon_0 \varepsilon' E^2 + \mu_0 \mu' H^2) \]  

\[
\text{Where P is power f is the frequency of the microwave, E is the electric field because of microwave, } \varepsilon_0 = 8.8 \times 10^{-14}, \varepsilon' \text{represent the relative dielectric constant due to material and solvent, } \mu_0 = 4\pi \times 10^{-7} \text{ H/m, } \mu' \text{ represent relative permeability of the material and H is the magnetic field. Our materials are intrinsically nonmagnetic in nature; therefore, the second term isn't significant. So, the power absorbed by the material is given by}
\]

\[
P = 2\pi f (\varepsilon_0 \varepsilon' E^2) \]  

\[
\text{GBNM hybrid material dielectric constant is } \varepsilon' = 6.8 \text{ at 2.45 GHz obtained from experimental measurement as shown in Fig.S25 for GBNS, GBNH, and GBNM samples.}
electric measurement for GBNS, GBNM, and GBNH samples. The light green oval shape represents the interest area.

The distribution of the electric field inside the microwave oven was obtained from the simulated result. As shown in Fig. S26[28]
Using dielectric constant ~ 6.8 obtained from experimental dielectric measurement for GBNM hybrid and electric field value ~2 x10^2 to 4x10^4 V/m\textsuperscript{[29-30]} obtained from numerous simulation results, in equation (3) and calculated absorbed power for our materials, it turns out to be ~ 0.38 eV/atom. When the microwave is brisked in a pulsed manner of ~ 60 s, the global temperature inside the reactor where DMF is present can go up to ~200 °C within a very short duration of time, ~ 60s \textsuperscript{[31]}, which will cause additional pressure ~0.5MPa inside the microwave reactor. The energy because of pressure built up inside the reactor is ~ 0.41 eV. Hence the adequate energy received by atoms within the microwave oven is ~ 0.41+0.38= 0.79 eV/ atom.

The skin depth is obtained by using the subsequent formula \textsuperscript{[32]}

\[
d = \frac{3.32 \times 10^7}{\omega \sqrt{\varepsilon_0 \varepsilon_r \tan(\varepsilon''/\varepsilon_r)}} \quad \text{(4)}
\]

Here \(\omega\) is the frequency of the microwave, \(\varepsilon'\)=6.8 is the real part, and \(\varepsilon''\)=2.55 is the imaginary part of the dielectric constant at 2.45GHz in the case of the GBNM sample. Substituting all values in equation (4) to obtain penetration depth, it turns out to be ~\(d=11.47\) mm. Thus, our material exhibited low loss, which is suitable for better microwave absorption and minimal conversion of microwave energy into heat energy.

The bond formation energies because of varied bonds C-N ~ 0.32 eV, B-C ~1.62 eV, \textsuperscript{[33]} Graphitic N ~0.06 eV, Graphitic N2 ~ 0.173 eV \textsuperscript{[34]}, nitrogen and boron bonded together with carbon atoms ~ 0.168 eV, 2 nitrogen bonded with graphene ~1.69 eV and 2 boron bonded with graphene ~1.53 eV \textsuperscript{[35]}. Our synthesis techniques Heat mediated, solvothermal, and microwave-based hybridization techniques offer the energies ~ 0.05 eV, 0.46 eV, and 0.916 eV/atoms that are sufficient for bond formation.
Fig. S27  Specific heat variation with temperature for RGO, BN, GBNS, **GBNH** and GBNS samples.
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