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1 **Enhanced channel mobility of hexagonal boron nitride/hydrogen-terminated diamond**  
2 **heterojunction field-effect transistor**

3 Yosuke Sasama,<sup>1, a)</sup> Takuya Iwasaki,<sup>2</sup> Masataka Imura,<sup>3</sup> Kenji Watanabe,<sup>3</sup>  
4 Takashi Taniguchi,<sup>2</sup> and Yamaguchi Takahide<sup>2,4</sup>

5 <sup>1)</sup>*International Center for Young Scientists, National Institute for Materials Science,*  
6 *Tsukuba 305-0044, Japan*

7 <sup>2)</sup>*Research Center for Materials Nanoarchitectonics, National Institute for Materials*  
8 *Science, Tsukuba 305-0044, Japan*

9 <sup>3)</sup>*Research Center for Electronic and Optical Materials,*  
10 *National Institute for Materials Science, Tsukuba 305-0044,*  
11 *Japan*

12 <sup>4)</sup>*University of Tsukuba, Tsukuba 305-8571, Japan*

13 Hydrogen-terminated diamond field-effect transistors (FETs) using a hexagonal  
14 boron nitride (h-BN) gate insulator were fabricated on a diamond surface with re-  
15 duced surface roughness in the direction of source/drain electrodes. The diamond  
16 surface was prepared on a mesa structure using chemical vapor deposition (CVD)  
17 with a low methane concentration. The hydrogen-terminated surface was laminated  
18 with the h-BN gate insulator without air exposure to prevent the adsorption of at-  
19 mospheric surface acceptors. The hydrogen-terminated diamond FET exhibited a  
20 high mobility of  $\approx 1000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  at room temperature. We performed theoret-  
21 ical analysis on the temperature and carrier density dependences of mobility, which  
22 suggested that Coulomb and surface roughness scattering were effectively reduced.  
23 The high mobility obtained in this study indicates the high potential of diamond as  
24 a semiconducting material. This study can contribute to the future development of  
25 diamond devices.

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<sup>a)</sup>SASAMA.Yosuke@mims.go.jp

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26 Diamond is a wide-gap semiconductor with a high breakdown electric field, high thermal  
27 conductivity, and high mobility, making it advantageous for power device applications<sup>1,2</sup>.  
28 Several types of diamond field-effect transistors (FETs) have been developed, including  
29 Schottky metal-semiconductor FETs<sup>3</sup>, junction FETs<sup>4</sup>, and deep-depletion FETs<sup>5</sup>. One of  
30 the most widely developed FETs is the hydrogen-terminated diamond FET, in which the  
31 diamond surface is terminated with hydrogen<sup>6</sup>. This is because terminating the diamond  
32 surface with hydrogen reduces the surface state density, and the higher energy of the valence  
33 band maximum facilitates the induction of hole carriers<sup>7</sup>.

34 Hydrogen-terminated diamond FETs often use the surface conductivity induced by trans-  
35 fer doping.<sup>8</sup> The surface conductivity can be obtained simply by exposing the hydrogen-  
36 terminated surface to air.<sup>9</sup> Additionally, carrier density can be increased to  $>10^{14}$   $\text{cm}^{-2}$  by  
37 exposure to  $\text{NO}_2$  gas<sup>10</sup> or the deposition of oxides with a large electron affinity<sup>11</sup>.

38 However, negative charges associated with the formation of surface conductivity on the  
39 hydrogen-terminated surface (ionized surface acceptors or atmospheric negative charges)  
40 scatter carriers and reduce the mobility of FETs. Reduced mobility is undesirable for high-  
41 speed and low-loss operation of FETs. To increase the mobility of diamond FETs, we  
42 have fabricated FETs with a reduced surface acceptor density using a hexagonal boron  
43 nitride (h-BN) gate insulator<sup>12-14</sup> and an air-free process<sup>15</sup>, in which the hydrogen-terminated  
44 surface is not exposed to air. We have obtained a high mobility of  $680 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  with a  
45 relatively high carrier density of  $6.6 \times 10^{12} \text{ cm}^{-2}$ . In addition to the high mobility, the  
46 FETs also demonstrate normally off operation, low on-resistance, and high on-off ratio  
47 simultaneously.<sup>15</sup>

48 In this study, we made two primary improvements to further increase the mobility of  
49 diamond FETs. The first is the reduction of surface roughness on a diamond. Surface  
50 roughness can contribute to the formation of surface states<sup>16</sup>. Charges trapped at surface  
51 states cause Coulomb scattering, resulting in reduced mobility. Mobility is also reduced by  
52 surface roughness scattering. Here, we grew a relatively flat diamond film on a mesa struc-  
53 ture using chemical vapor deposition (CVD) with a low methane concentration<sup>17</sup>, and we  
54 fabricated FETs on the diamond film. The second improvement is the lamination technique  
55 of h-BN. Here, we use pick-up method to laminate h-BN. The use of this pick-up method<sup>18,19</sup>  
56 effectively avoids the formation of contamination bubbles, which are often problematic when  
57 creating van der Waals heterostructures. Through the use of a curvature polymer and slowly

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58 laminating the layers, bubbles are pushed out, resulting in the formation of a clean interface  
59 free from contamination bubbles. Therefore, this method has been increasingly employed in  
60 recent years to fabricate van der Waals heterostructures<sup>20</sup>, including recent h-BN/diamond  
61 hetero devices.<sup>21,22</sup>

62 First, we focused on the growth of diamond films. Generally, a diamond (111) substrate  
63 has a misorientation angle, and atomic steps exist on the substrate surface. When diamond is  
64 grown using CVD with a low methane concentration on a (111) diamond substrate, it grows  
65 laterally from atomic steps, and atomically flat diamond can be formed<sup>17</sup>. Following the  
66 previous report, we grew diamond films. We used type-Ib (111) single-crystalline diamond  
67 substrate manufactured by Sumitomo Electric Industries. After the mesa structure was  
68 formed on the diamond substrate using oxygen plasma etching, the diamond was grown  
69 using CVD under the following conditions: hydrogen flow rate of 1000 sccm, methane flow  
70 rate of 0.1 sccm, microwave power of 800 W, and pressure of 80 torr. The quality of the  
71 diamond surface was evaluated using an atomic force microscope (AFM). Measurements  
72 were performed on an area of 20  $\mu\text{m}$  square. Steps existed in the direction perpendicular  
73 to the [111] direction, but a relatively flat diamond film with an average surface roughness  
74 (Ra) of  $\approx 0.15$  nm was grown (Fig. 1c).

75 Subsequently, we fabricated an FET using a h-BN gate insulator with a Hall-bar structure  
76 (Fig. 1a) on the CVD-grown diamond. First, Ti/Pt was deposited as electrodes. Here, the  
77 source/drain electrodes (current electrodes in the Hall bar) were placed parallel to the step  
78 of the diamond. An annealing was performed in a hydrogen atmosphere (hydrogen flow  
79 rate of 500 sccm, pressure of 80 torr) at 650°C for 35 min to form TiC, which functions as  
80 an ohmic electrode. Thereafter, the diamond surface was exposed to hydrogen plasma to  
81 remove resist residue and atmospheric adsorbates. The hydrogen plasma conditions were as  
82 follows: hydrogen flow rate of 500 sccm, microwave power of 300 W, pressure of 30 torr,  
83 and duration of 10 min. Annealing and hydrogen plasma treatment were also performed in  
84 another CVD chamber under the same conditions as in the first chamber. The second CVD  
85 chamber could be connected to a vacuum suitcase. The diamond was vacuum-transferred  
86 to an Ar-filled glove box using the vacuum suitcase to avoid the adsorption of atmospheric  
87 adsorbates on the hydrogen-terminated diamond surface. In the glove box, the hydrogen-  
88 terminated diamond was laminated with a cleaved h-BN thin film. Here, we used the pick-up  
89 method<sup>18,19,21,22</sup> for the h-BN transfer. A single-crystalline h-BN was cleaved with scotch

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90 tape and transferred onto a Si substrate. The cleaved h-BN film was then picked up from  
91 the Si substrate using a polymer stamp and transferred onto the diamond. The lamination  
92 of diamond with h-BN was performed at room temperature and then the diamond was  
93 heated to 110°C to release the h-BN from the stamp. The thickness of the h-BN film used  
94 in this study was 16 nm. After the h-BN lamination, the diamond substrate was annealed  
95 in the glove box at 200°C for 3 h. The diamond substrate was then removed from the glove  
96 box. The h-BN gate insulator was etched into a Hall-bar shape using capacitively coupled  
97 plasma reactive ion etching (RIE) with N<sub>2</sub>, CHF<sub>3</sub>, and O<sub>2</sub> gases. The diamond surface,  
98 except for the area underneath the h-BN, was converted to oxygen termination during this  
99 process, providing device isolation. Subsequently, Ti/Au/Ti films were deposited on the h-  
100 BN as a gate electrode. The hydrogen-terminated diamond may attract negatively charged  
101 impurities, resulting in the intercalation of impurities at the interface between the diamond  
102 and h-BN, because the hydrogen-terminated diamond surface is slightly positively charged  
103 owing to the difference in electronegativity between carbon and hydrogen atoms<sup>8</sup>. To prevent  
104 impurities from contaminating the interface, we deposited 30-nm Al<sub>2</sub>O<sub>3</sub> using atomic layer  
105 deposition. We consider that the Al<sub>2</sub>O<sub>3</sub> passivation contributes to the long-term stability  
106 of the device. Thereafter, through holes were opened in Al<sub>2</sub>O<sub>3</sub> using wet etching with  
107 dilute tetramethylammonium hydroxide. Finally, the gate contact was fabricated through  
108 the sputter deposition of Ti/Au. The optical microscope image of the fabricated device is  
109 shown in Fig. 1b.

110 We evaluated the electrical characteristics of the FET. Figure 2a shows the transfer  
111 characteristics. (See also Fig. S1 of the supplementary material.) The FET exhibits p-type  
112 FET characteristics. The maximum sheet conductance is 1.1 mS, which corresponds to a  
113 sheet resistance of 0.93 kΩ. The sheet conductance ( $\sigma$ ) is obtained as  $\sigma = (L_p/W_G)(1/R)$ ,  
114 where  $R$  is the resistance measured using the four-terminal method,  $W_G$  ( $= 0.97 \mu\text{m}$ ) is the  
115 gate width, and  $L_p$  ( $= 3.0 \mu\text{m}$ ) is the distance between the voltage probes. This device  
116 has a fringe region in which the diamond surface is hydrogen-terminated and covered by  
117 h-BN, but it is not under the gate electrode. This region may be conductive owing to hole  
118 carriers generated by surface acceptors originating from residual gases in the glove box. The  
119 conductance in the fringe region may result in an overestimate of the sheet conductance.  
120 However, if we assume that the sheet conductance in the fringe region is equal to that in the  
121 gate-overlapped region at zero gate voltage, the impact of the fringe region on the maximum

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122 sheet conductance is estimated to be only 7%. This value is obtained as  $\Delta\sigma = \sigma(V_{GS} =$   
 123  $0) \times (W_{h-BN} - W_G)/W_{h-BN}$  using the h-BN width ( $W_{h-BN} = 3.97 \mu\text{m}$ ) and gate width.  
 124 The threshold voltage evaluated from the transfer curve is 0.67 V. Unlike our previous  
 125 study, the threshold voltage is positive, which indicates normally on operation. We consider  
 126 that the reason for this positive threshold voltage is a lower concentration of nitrogen<sup>23</sup>  
 127 acting as a donor unlike in the previous one<sup>15</sup>. The diamond substrates used in our previous  
 128 study were type-IIa high pressure, high temperature(HPHT)-grown diamond purchased from  
 129 the Technological Institute for Superhard and Novel Carbon Materials (TISNCM), and  
 130 secondary ion mass spectrometry (SIMS) on a similar substrate showed that the nitrogen  
 131 concentration in the substrate was  $\approx 500$  ppb. In contrast, the nitrogen concentration in  
 132 diamond grown under the similar conditions as in the present study was below the detection  
 133 limit of SIMS measurement ( $< 20$  ppb).

134 Figure 2b shows the output characteristics. The drain-source length is  $8.8 \mu\text{m}$ . The  
 135 maximum drain current density is  $\approx 240 \text{ mA mm}^{-1}$ . The drain current density is obtained  
 136 by dividing the drain current by the gate width. Based on a simple calculation considering  
 137 the gate and h-BN width, the current flowing through the fringe region ( $I(V_{GS} = 0) \times$   
 138  $((W_{h-BN} - W_G)/W_{h-BN})$ ) is  $15 \text{ mA mm}^{-1}$ , which is as low as 6% of the maximum drain  
 139 current density. The FET is not completely turned off because of the fringe current. Because  
 140 the gate electrode does not cover the fringe region, the hole carriers do not deplete even when  
 141 a positive gate voltage is applied. Therefore, a small drain current flows even when a positive  
 142 gate voltage is applied. To completely turn off the FET, we believe that device isolation  
 143 techniques should be improved.

144 Carrier density and mobility were evaluated using Hall-effect measurements. Figure 2c  
 145 shows the gate voltage dependence of the carrier density. The carrier density increased  
 146 monotonically, with a maximum carrier density of  $4.7 \times 10^{12} \text{ cm}^{-2}$ . Figure 2d shows the Hall  
 147 mobility. Field-effect mobility ( $\mu_{FE} = \frac{t_{hBN}}{\epsilon_{hBN}} \left| \frac{\partial \sigma}{\partial V_{GS}} \right|$ ) and effective mobility ( $\mu_{eff} = \frac{t_{hBN}}{\epsilon_{hBN}} \frac{\sigma}{|V_{GS} - V_{th}|}$ )  
 148 estimated from the slope of the transfer characteristic (Fig. 2a) are also shown.  $\sigma$  is the sheet  
 149 conductance shown in Fig. 2a,  $V_{GS}$  is the gate voltage,  $t_{hBN}$  is the thickness of h-BN, and  
 150  $\epsilon_{hBN} = 3$  is the dielectric constant of h-BN. For the estimation of Hall mobility, the conduc-  
 151 tance of the side paths was subtracted using the same method as in our previous work<sup>12</sup>. The  
 152 Hall mobility ( $\mu_{Hall}$ ) is calculated as  $\mu_{Hall} = (\sigma - \sigma(V_{GS} = 0))(W_{h-BN} - W_G)/W_{h-BN}/(n_H e)$ .  
 153  $\sigma$  is the sheet conductance,  $n_H$  is the Hall carrier density, and  $e$  is the elementary charge

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154 (Figure S2 of the supplementary material). A high mobility of  $\approx 1000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  was ob-  
155 served in the Hall, field-effect, and effective mobilities. The Hall mobility is slightly higher  
156 than effective and field-effect mobilities, which might be related to the current paths besides  
157 the channel. We note that we can rule out the possibility that the high mobility originates  
158 from boron contamination in the diamond film. The density of carriers induced by boron  
159 does not increase with increasing negative gate voltage; hence, at least the field-effect mo-  
160 bility is considered to be the mobility of a two-dimensional hole gas induced by applying a  
161 gate voltage on a hydrogen-terminated diamond.

162 We also evaluated the temperature dependence of the mobility. Figure 3 shows the tem-  
163 perature dependence of the mobility evaluated using Hall-effect measurements. The mobility  
164 of our present FETs fabricated on a CVD-grown diamond increases with decreasing temper-  
165 ature more rapidly than our previous FETs fabricated on a diamond surface without CVD  
166 growth. The maximum mobility is  $\approx 2300 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$  at 100 K. Below 100 K, the mobility  
167 could not be evaluated accurately owing to the large contact resistance at low temperature.  
168 The large contact resistance at low temperatures may originate from the gap formed at the  
169 h-BN/diamond interface near the Hall-bar electrodes, as discussed in our recent paper<sup>22</sup>.  
170 The cleaner surface compared with before may increase the electrical resistance of diamond  
171 underneath the gap near electrodes and, therefore, the contact resistance at low tempera-  
172 tures. We consider that the use of gate electrode self-aligned with source/drain electrodes<sup>22</sup>  
173 can eliminate the gap formed at the h-BN/diamond interface near the electrodes and reduce  
174 the contact resistance.

175 The reduction of Coulomb and surface roughness scattering was supported by the the-  
176 oretical analysis of mobility. Figure 4 shows the theoretical analysis of (a) carrier density  
177 dependence and (b) temperature dependence of mobility. (See also Fig. S3 of the supple-  
178 mentary material.) The mobility was calculated theoretically using the same method as  
179 in our previous report (Supplementary Information in Ref.<sup>15</sup>). As shown in Fig. 4, the  
180 theoretical calculations of mobility adequately explain the experimental results for both the  
181 carrier density and the temperature dependences of mobility. Here, the boron and nitrogen  
182 concentrations were set to 1 and 5 ppb, respectively, and the average roughness ( $\Delta$ ) was set  
183 to 0.15 nm based on AFM measurements. We have tuned the interfacial charge density ( $n_{ic}$ )  
184 and distance between the interfacial charges and hole carriers ( $d$ ). The values of  $d$  and  $n_{ic}$   
185 were determined by minimizing the total sum of squared residuals between the calculated

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186 and experimental mobilities at  $V_{GS} = -2, -4$  and  $-6$  V. Consequently, the optimal values  
 187 were found to be  $d = 0.0$  nm and  $n_{ic} = 2.4 \times 10^{11}$  cm $^{-2}$ . The obtained value of  $d$  is con-  
 188 sistent with those assumed in references<sup>14, 15</sup>, and<sup>25</sup>, and is similar to the value reported in  
 189 reference<sup>24</sup>. The experimental results of our previous study<sup>15</sup> are well explained when the  
 190 calculation parameter values are  $n_{ic} = 6 \times 10^{11}$  cm $^{-2}$ ,  $\Delta = 0.18$  nm, whereas the results  
 191 of this study are well explained when the calculation parameter values are  $n_{ic} = 2.4 \times 10^{11}$   
 192 cm $^{-2}$ ,  $\Delta = 0.15$  nm. The reduction in  $n_{ic}$  is considered to be owing to both the decrease in  
 193 the surface roughness and improvement in the h-BN lamination technique. The reduction in  
 194 surface roughness is expected to decrease the density of the interface states. The use of the  
 195 pick-up method for the h-BN lamination has also contributed to the improvement in mo-  
 196 bility by reducing the charged impurity density at the hydrogen-terminated diamond/h-BN  
 197 interface.

198 In summary, we fabricated diamond FETs using an h-BN gate insulator on a diamond  
 199 film grown using CVD with a low methane concentration. Through the CVD with a low  
 200 methane concentration, a diamond film with reduced surface roughness in the direction of  
 201 source/drain electrodes was grown. The h-BN gate insulator was formed via the cleavage of  
 202 h-BN and a pick-up method using a curvature polymer stamp. We demonstrated that a high  
 203 hole mobility of  $\approx 1000$  cm $^2$ V $^{-1}$ s $^{-1}$  can be achieved at room temperature in the hydrogen-  
 204 terminated diamond FET by improving the quality of the interface between the hydrogen-  
 205 terminated diamond and gate insulator. The theoretical analysis of the carrier density and  
 206 temperature dependences of mobility supported that this high mobility was achieved by re-  
 207 ducing surface roughness and interface charge density. This mobility approaches that of the  
 208 electron mobility in GaN high electron mobility transistors (HEMTs), suggesting that com-  
 209 bining our diamond FET with a GaN-HEMT can enable high-performance complementary  
 210 circuits based on wide-bandgap semiconductors.

## 211 Supplementary Material

212 See the supplementary material for additional analysis on the FET's long-term electrical  
 213 stability, calculation of Hall mobility accounting for fringe conductance, and the theoretical  
 214 and experimental exploration of temperature-dependent mobility at various gate voltages.

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215 **Data availability**

216 The data supporting the findings of this study are available from the corresponding author  
217 upon reasonable request.

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<sup>223</sup> search Infrastructure for Materials and Nanotechnology in Japan (ARIM) (Proposal No.  
<sup>224</sup> JPMXP1224NM5213).

<sup>225</sup> **Competing Interests**

<sup>226</sup> The authors declare that they have no competing financial interests.

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<sup>227</sup> **Data availability**

<sup>228</sup>     The data that support the findings of this study are available from the corresponding  
<sup>229</sup> author upon reasonable request.

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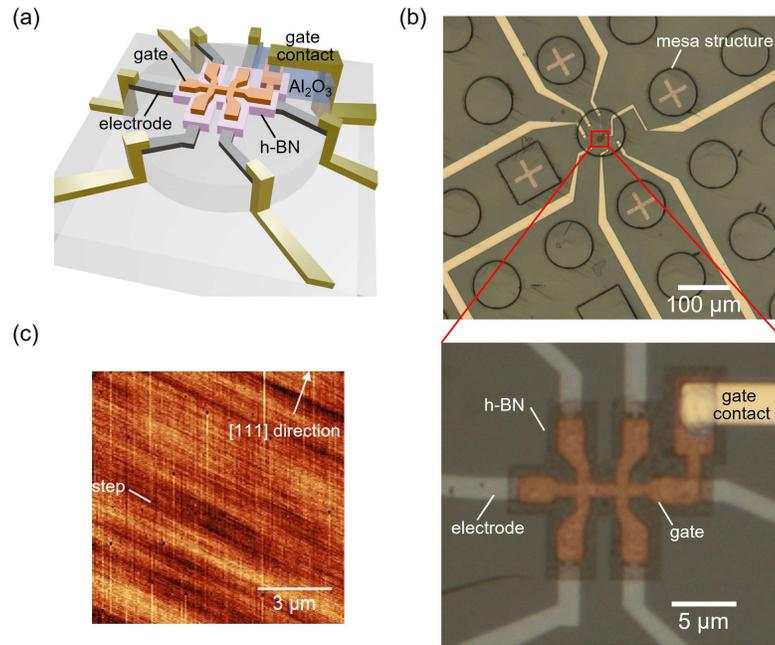


FIG. 1. (a) Schematic and (b) optical microscope image of the diamond FET fabricated in this study. (c) AFM image of the diamond surface after CVD growth. The arrow represents the projection of the [111] direction onto a two-dimensional plane.

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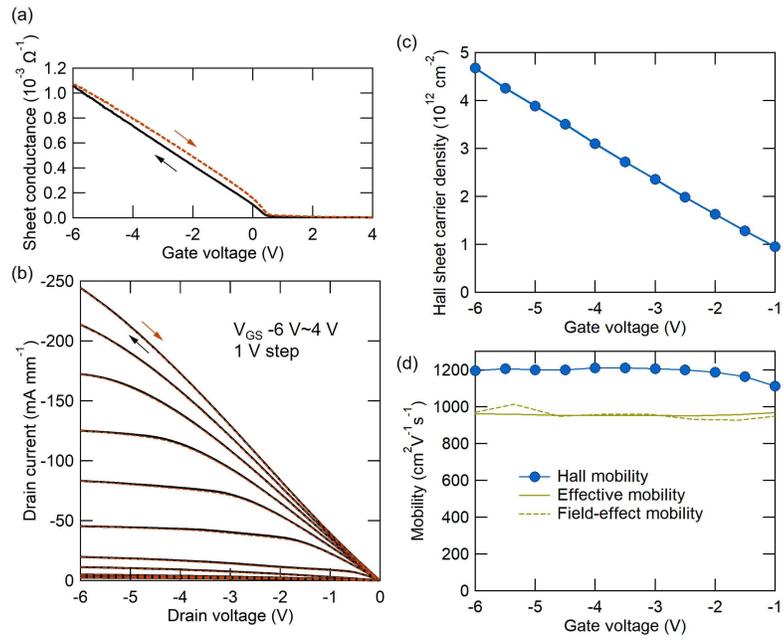


FIG. 2. Device characteristics of the diamond FET (Sample B1) at 300 K. (a) Transfer characteristics, (b) output characteristics, (c)(d) gate voltage dependence of (c) carrier density and (d) mobility.

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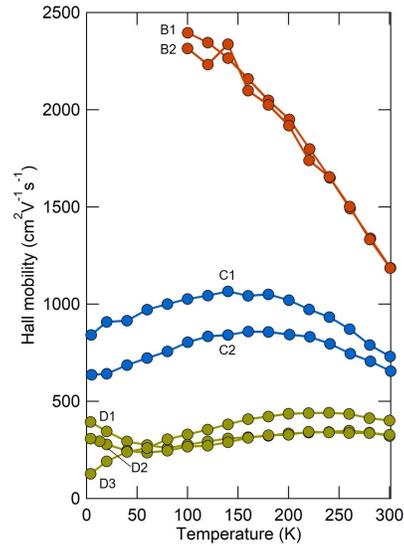


FIG. 3. Temperature dependence of Hall mobility. The points connected by lines are the measurement results of the same device measured at different temperatures. Samples B1 and B2 were fabricated on a CVD-grown diamond surface, whereas samples C1, C2 (ref.<sup>15</sup>) and D1-D3 (ref.<sup>12</sup>) were fabricated on a diamond surface without CVD growth. Samples B1, B2, C1, and C2 were fabricated without air exposure to hydrogen-terminated surface, whereas samples D1-D3 were fabricated on an air-exposed hydrogen-terminated surface. Measurements were performed with the application of a gate voltage ( $-6$  V for B1,  $-15$  V for B2,  $-8$  V for C1,  $-10$  V for C2,  $-4$  V for D1,  $-7$  V for D2,  $-5$  V for D3, depending on the thickness of h-BN). Sample B2 exhibited fluctuations in electrical measurements at low temperatures owing to a high contact resistance. Sample B2 was fabricated using a process in which the gate electrode was self-aligned with source/drain electrodes. Some of the hydrogen-terminated surface in the access region was damaged during the h-BN etching process, resulting in high contact resistance in sample B2.

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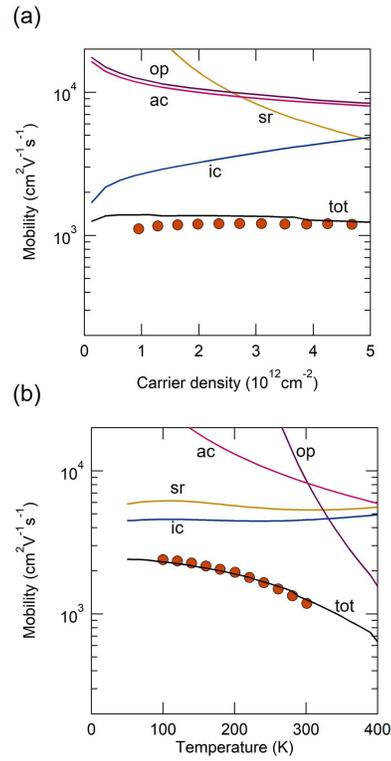


FIG. 4. Theoretical analysis of mobility. (a) Carrier density dependence and (b) temperature dependence of mobility. Red circles show experimental results for sample B1. The carrier density dependence was measured at 300 K. The temperature dependence was measured with an applied gate voltage of  $-6$  V. Solid lines show the results of the theoretical calculation of mobility. The labels “ic,” “sr,” “ac,” and “op” indicate the calculated mobilities limited by interface charges, surface roughness, acoustic phonon, and optical phonon, respectively. The mobility limited by background impurity scattering has values much higher than the plot range. The label “tot” indicates the calculated mobility considering all the above scattering.