Supporting Information

Giant Piezoresistive Gauge Factor in Vein-membrane/Graphene Sensors with a Wide Linear Working Range

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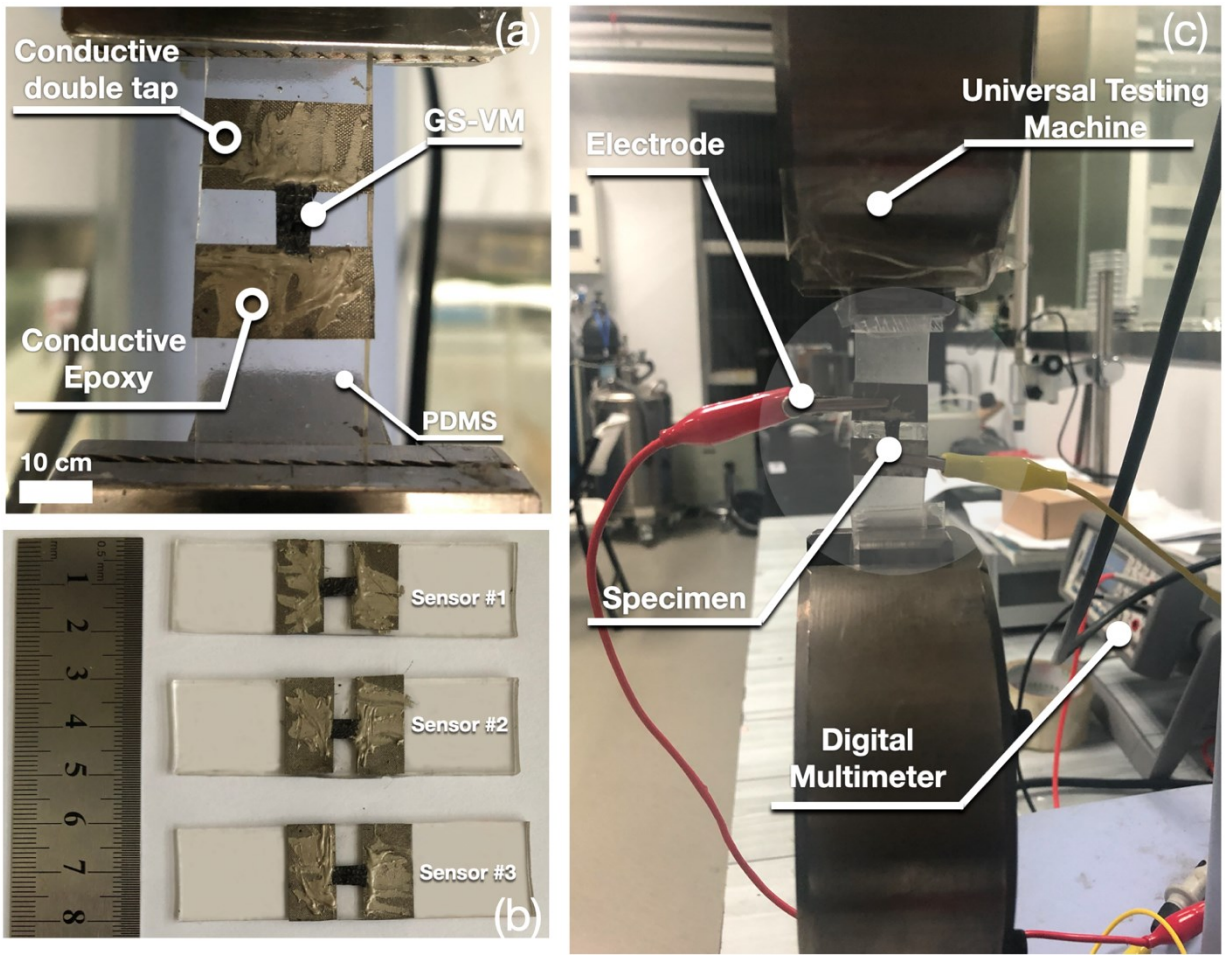

Figure S1. (a) and (b) GS-VM piezoresistive sensors; (c) testing method of the specimen.

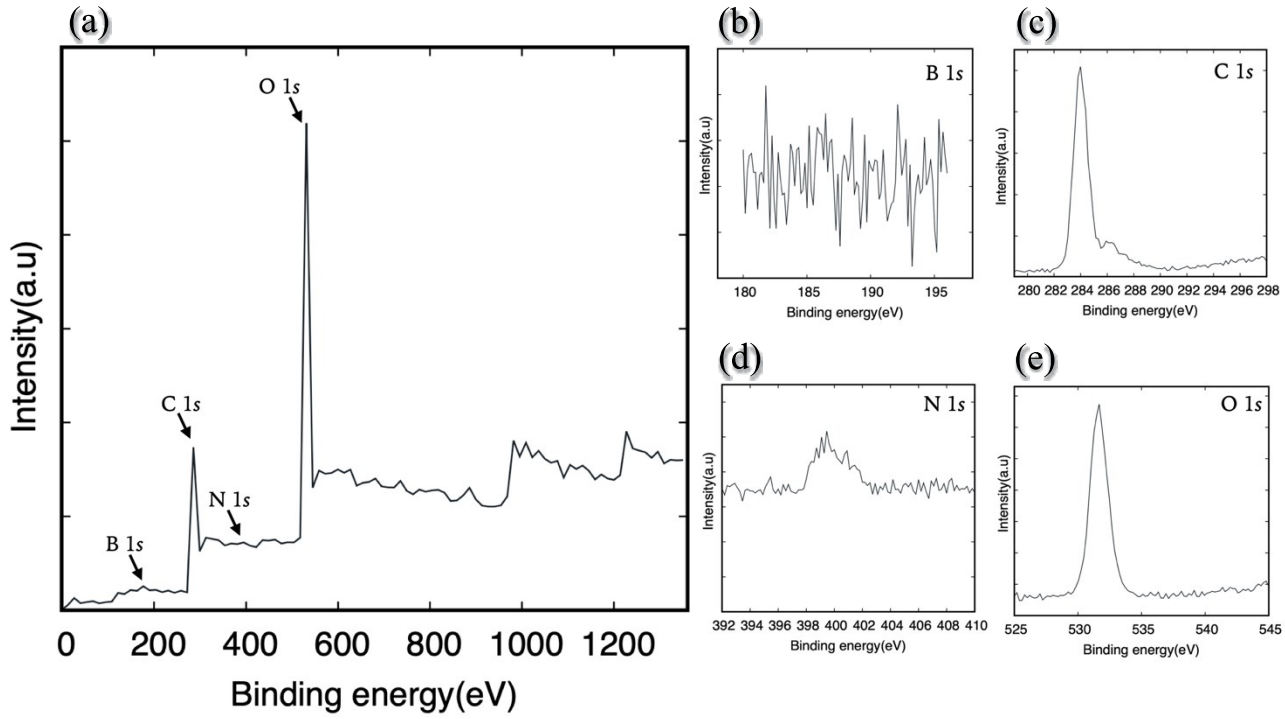

Figure 2s. (a-e) XPS of the vein of the dead leaf presents the overall analysis, B 1*s*, C 1*s*, N 1*s*, and O 1*s*, respectively.

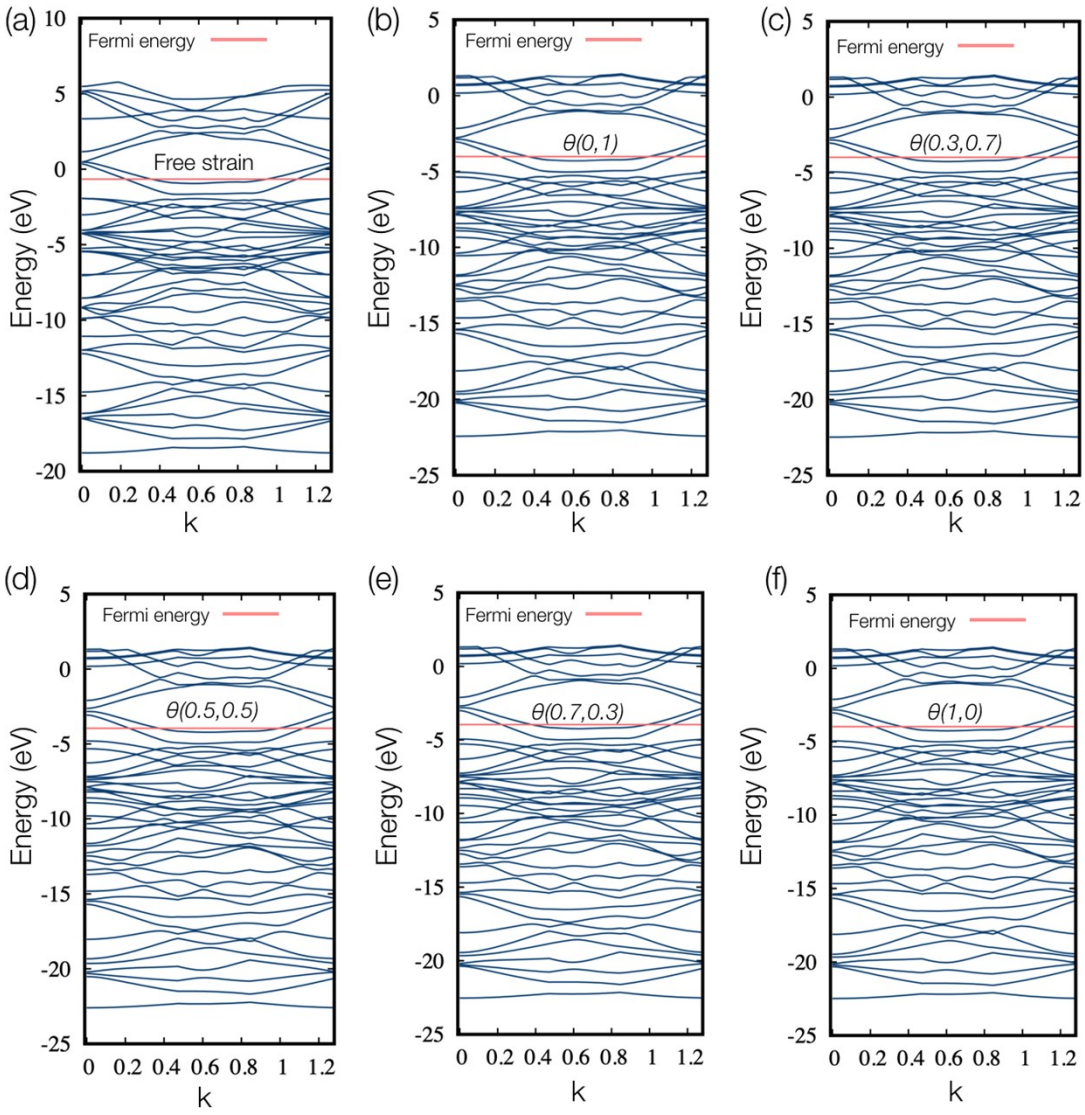

Figure S3. The electronic energy band; (a) the energy band in the strain-free state; (b-f) the energy band when subjected to 1% mechanical strain along different θ-directions.

Appendix A

To qualitatively estimate the equivalent resistance of GS-VM, we assumed that the number of resistors at every level, i.e., $s[L(k)]$, mainly depended on the fractal feature at each level. k (1,2,3, ..., n-1, n, ...) represents a certain level of the GS-VM, and $s[L(k)]$ is the number of resistors. Therefore, the equivalent resistance of a certain level can be expressed by Equation (A.1) due to the self-similar nature of fractals [1].

$$\frac{1}{R_{L(k)}} = \frac{s[L(k)]}{\lambda^k R_{L(0)}}$$

(A.1)

Thus, the total equivalent resistance of GS-VM can be expressed by Equation (A.2).

$$R = \sum_{k=0}^{\infty} \frac{\lambda^k}{s[L(k)]} R_{L(0)} \tag{A.2}$$

where λ represents the scalar of this fractal structure, $\lambda \in (0,1)$, and the resistance of the first top-level is $R_{L(0)}$. If we assume that $s[L(k)] = k$, then the total equivalent resistance of the GS-VM can be computed in Equation (A.3) when k goes to infinity.

$$R = R_{L(0)} log(\frac{1}{1-\lambda})$$

(A.3)

Equation (A.3) implies if the number of levels in the multi-level network goes to infinity, then the total resistance can be converged. This means that the GS-VM would not change its total equivalent resistance with a more complex fractal structure, but it will improve its ability to experience large deformations. Therefore, the GS-VM shows satisfactory repeatability.

Appendix B

The well-known relation is presented in Equation (B.1), where R, l, S, and ρ denote the resistance, length, crossing-area, and resistivity, respectively.

$$R = \frac{l}{S}\rho \tag{B.1}$$

Hence, GF is defined as:

$$GF = \frac{1^{R - R_0}}{\varepsilon \quad R_0}$$

(B.2)

Here, the subscript 0 represents the strain-free state, and ε is the applied strain. If the geometric parameters are ignored, substituting Equation (B.1) into Equation (B.2), Equation (B.2) can be expressed as:

$$GF = \frac{1}{\varepsilon}(\frac{\rho}{\rho_0} - 1) \tag{B.3}$$

However, the carriers, i.e., electrons and holes, play an important role in conductivity (reciprocal resistivity). Hence, the conductivity can be expressed as:

$$\sigma = nq\mu_e + pq\mu_h \tag{B.4}$$

where μ_e and μ_h are the electron and hole mobilities, respectively, and q is the elementary electrical charge.

$$\mu_e = \frac{q\tau_e}{m_e^*} \tag{B.5a}$$

and

$$\mu_h = \frac{q\tau_h}{m_h^*} \tag{B.5b}$$

where m^* is the effective mass, τ is the relaxation time, and the subscripts e and h represent electrons in the conduction band and holes in the valence band, respectively. Furthermore,

$$\frac{1}{m^*} = \frac{1}{\hbar^2} \frac{\partial^2 \epsilon}{\partial k \partial k} \tag{B.6}$$

where $\hbar$ is Planck's constant, and $\boldsymbol{k}$ is the wavenumber vector. In Equation (B.4), n and p represent the concentrations of electrons and holes, respectively. The concentrations of electrons can be calculated by:

$$n = \int_{\epsilon_c}^{\infty} D_e(\epsilon) f(\epsilon) d\epsilon \tag{B.7}$$

and

$$p = \int_{-\infty}^{\epsilon_v} D_h(\epsilon)[1 - f(\epsilon)] d\epsilon \tag{B.8}$$

where $f(\epsilon)$ is the Fermi-Dirac distribution, which is given by:

$$f(\epsilon) = \frac{1}{exp[(\epsilon - \epsilon_F)/k_B T] + 1} \tag{B.9}$$

where $D(\epsilon)$, ϵ, ϵ_F, k_B, and T represent the density of states, the energy of the electronic band, Fermi energy, Boltzmann's constant, and temperature, respectively. Equations (B.4)-(B.9) were introduced by Kittel [2]. Therefore, the electrical conductivity can be summarized as:

$$\sigma = \frac{q^2 \tau_e}{\hbar^2} \int_{\epsilon_c}^{\infty} \frac{\partial^2 \epsilon}{\partial k \partial k} D_e(\epsilon) \frac{1}{\exp\left[\frac{\epsilon - \epsilon_F}{k_B T}\right] + 1} d\epsilon + \frac{q^2 \tau_h}{\hbar^2} \int_{-\infty}^{\epsilon_v} \frac{\partial^2 \epsilon}{\partial k \partial k} D_e(\epsilon) \left[1 - \frac{1}{\exp\left[\frac{\epsilon - \epsilon_F}{k_B T}\right] + 1}\right] d\epsilon \tag{B.10}$$

The subscripts c and v represent the conduction and valence bands, respectively. First, all relaxation times were considered to be constant to conveniently use Gamil's idea [3]. The right sides of Equations (B.6)-(B.8) were then discretized to obtain:

$$\frac{1}{m^*} = \frac{1}{\hbar^2} \frac{\epsilon(k + 2\Delta k) + \epsilon(k) - 2\epsilon(k + \Delta k)}{\Delta k^2} \tag{B.11}$$

$$n_i = \frac{1}{A}\sum_{i=j+1}^{\infty} w_{k_i} f[\epsilon(k_i)] \tag{B.12}$$

$$p_i = \frac{1}{A}\sum_{i=0}^{j} w_{k_i}(1 - f[\epsilon(k_i)]) \tag{B.13}$$

where index i represents the number of sub-bands in the electronic energy band structure, and *j* is the number of the valence band. A is the area of the boron-doped graphene supercell. w_{k_i}, calculated by Newton-Cotes integration[4], is the weight coefficient of k. Here *k* represents the wavenumber along the stretch direction. Combining Equation (B.3) and Equations (B.11)-(B.13), the GF at the atomic scale can, therefore, be given in Equation (3).

Appendix C

The stretch direction can be defined by Equation (C.1), where *u* and *v* represent the vector coordinates of stretching in the Cartesian system.

$$\theta(u,v) = atan(\frac{u}{v}) \tag{C.1}$$

a and ***b*** can be expressed by Equations (C.2a)-(C.2b) in the g_j-system.

$$a = a^i g_i \tag{C.2a}$$

$$b = b^i g_i \tag{C.2b}$$

However,

$$g_j = Fe_j \tag{C.3}$$

where F can be calculated by Equation (C.4) according to the crystal structure of the supercell.

$$F = v_0 \sin\left(\frac{\pi}{6}\right)\begin{bmatrix} \dfrac{\cos(\theta)}{\sin(\frac{\pi}{3}+\theta)} & \dfrac{\sin(\theta)}{\sin(\frac{\pi}{3}+\theta)} \\ -\dfrac{\sin(\theta)}{\sin(\frac{5\pi}{6}-\theta)} & \dfrac{\cos(\theta)}{\sin(\frac{5\pi}{6}-\theta)} \end{bmatrix} \tag{C.4}$$

Hence,

$$\begin{cases} g_1 = v_0 \sin\left(\frac{\pi}{6}\right)\left[\dfrac{\cos(\theta)}{\sin\left(\frac{\pi}{3}+\theta\right)}e_1 + \dfrac{\sin(\theta)}{\sin(\frac{\pi}{3}+\theta)}e_2\right] \\ g_2 = v_0 \sin\left(\frac{\pi}{6}\right)\left[-\dfrac{\sin(\theta)}{\sin\left(\frac{5\pi}{6}-\theta\right)}e_1 + \dfrac{\cos(\theta)}{\sin(\frac{5\pi}{6}-\theta)}e_2\right] \end{cases} \tag{C.5a}$$

$$F^{-1} = \frac{1}{v_0 \sin\left(\frac{\pi}{6}\right)}\begin{bmatrix} \cos(\theta)\sin(\frac{\pi}{3}+\theta) & -\sin(\theta)\sin(\frac{5\pi}{6}-\theta) \\ \sin(\theta)\sin(\frac{\pi}{3}+\theta) & \cos(\theta)\sin(\frac{5\pi}{6}-\theta) \end{bmatrix} \tag{C.5b}$$

In Equations (C.5a)-(C.5b), v_0 can be easily calculated via the crystal structure parameters in the strain-free state.

For convenience,

$$B = F^{-1} \tag{C.6}$$

Hence,

$$a = CBa_0 \tag{C.7a}$$

$$b = CBb_0 \tag{C.7b}$$

$\boldsymbol{C}$ can be represented by Equation (C.8a), while $\boldsymbol{a_0}$ and $\boldsymbol{b_0}$ are the crystal structure parameters in the Cartesian coordinate system, which can be calculated by Equations (C.8b)-(C.8c).

$$\boldsymbol{C} = \begin{bmatrix} 1+\frac{\varepsilon}{2} & 0 \\ 0 & 1-\frac{\varepsilon\nu}{2} \end{bmatrix} \tag{C.8a}$$

$$a_0 = (\frac{\sqrt{3}}{3}v_0, -v_0) \tag{C.8b}$$

$$b_0 = (\frac{\sqrt{3}}{3}v_0, v_0) \tag{C.8c}$$

In Equation (C.8a), ε and ν are applied strain and Poisson's ratio, respectively. ν was equal to 0.14 and 0.28 when the crystal structure underwent stretching along the zigzag- and armchair-directions, respectively [3]. In this paper, ν was calculated using linear interpolation when stretched in the zigzag- and armchair-directions in Equation (C.9)

$$\nu = 0.14u + 0.28v \tag{C.9}$$

Moreover,

$$G_{ij} = g_i \cdot g_j \tag{C.10a}$$

where G_{ij} represents the components of the metric tensor, whose matrix form can be obtained using Equation (C.10b):

$$G = [v_0 \sin\left(\frac{\pi}{6}\right)]^2 \begin{bmatrix} \frac{1}{\sin^2\left(\frac{\pi}{3}+\theta\right)} & 0 \\ 0 & \frac{1}{\sin^2\left(\frac{5\pi}{6}-\theta\right)} \end{bmatrix} \tag{C.10b}$$

Combining Equations (C.6)-(C.10b) and then substituting them into Equations (4a)-(4b), then the supercell parameters listed in Table 2 can be calculated.

Reference

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[2] Kittel, C., *Introduction to solid state physics* **2005**, John Wiley & Sons. *Inc., New York*.

[3] Gamil, M.; Nakamura, K.; El-Bab, A. M. F.; Tabata, O.; El-Moneim, A. A., Simulation of graphene piezoresistivity based on density functional calculations. *Modeling and Numerical Simulation of Material Science* **2013**, *3*(04), 117

[4] Stoer, J.; Bulirsch, R., *Introduction to numerical analysis* **2013**, (Vol. 12), Springer Science & Business Media.