Tunneling Magnetoresistance Transition and Highly Sensitive Pressure Sensors Based on Magnetic Tunnel Junctions with a Black Phosphorus Barrier

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ABSTRACT: Black phosphorus is a promising material to serve as a barrier for magnetic tunnel junctions (MTJs) due to weak van der Waals interlayer interactions. In particular, the special band features of black phosphorus may endow intriguing physical characteristics. Here we study theoretically the effect of band gap tunability of black phosphorus on the MTJs with the black phosphorus barrier. It is found that the tunneling magnetoresistance (TMR) may transition from a finite value to infinity owing to the variation in the band gap of black phosphorus. Combined with the latest experimental results of the pressure-induced band gap tunability, we further investigate the pressure effect of TMR in the MTJs with a black phosphorus barrier. The calculations show that the pressure sensitivity can be quite high under appropriate parameters. Physically, the high sensitivity originates from the TMR transition phenomenon. To take advantage of the high pressure sensitivity, we propose and design a detailed structure of highly sensitive pressure sensors based on MTJs with a black phosphorus barrier, whose working mechanism is basically different from that of conventional pressure sensors. The present pressure sensors possess four advantages and benefits: (1) high sensitivity, (2) good anti-interference, (3) high spatial resolution, and (4) fast response speed. Our study may advance new research areas for both the MTJs and pressure sensors.

INTRODUCTION

Black phosphorus is one type of two-dimensional semiconductor that has received tremendous attention owing to its two-dimensionality and the availability of advanced characterization techniques.1 For practical applications, black phosphorus has great potential in field effect transistors, photodetectors, batteries, etc.1 In the field of spintronics, weak van der Waals interlayer interactions make it possible for black phosphorus to serve as a barrier for magnetic tunnel junctions (MTJs). Up to now, research on MTJs with a phosphorus barrier has been rarely addressed.2 In particular, the band gap tunability of black phosphorus, which may play a key role in tunneling magnetoresistance (TMR), has never been adequately considered.

The band gap tunability of black phosphorus mainly manifests in two ways: thickness (layer) dependence and pressure dependence. On one hand, the band gap of black phosphorus is highly dependent on the thickness.1,3−5 For monolayer black phosphorus, the band gap is about 2 eV,1,3,5,6 while for bulk black phosphorus, the band gap is only about 0.3 eV.1,3,5,7 On the other hand, quite recently, S. Huang et al. investigated experimentally the pressure effect on the electronic structure of black phosphorus.8 They found that the relative changes in band gap versus pressure depend on the number of layers of black phosphorus, which can be expressed as follows:

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\[
\Delta E^N_g(P) = aP - \frac{\gamma_0}{2} \left( 1 + \frac{P}{P_{coh}} - 1 \right) \cos \left( \frac{1}{N+1} \pi \right)
\]

where \( P \) is the magnitude of the pressure, \( N \) is the number of the layers, \( a \) is the changing rate, \( \gamma_0 \) is the difference of overlapping integrals for the conduction band and valence band under 0 GPa, and \( P_{coh} \) is the cohesive pressure. In ref 8, both the experiments and the derivation of eq 1 are irrelevant to the substrate. That is, eq 1 represents the physical properties of black phosphorus alone. In addition, it is worth noting that eq 1 is valid only for the case where the thickness of black phosphorus is not greater than 50 nm. This is because, for bulk black phosphorus, there is additional in-plane compressive strain besides the normal strain.\(^8\) The above band gap limit, if \( \Delta \mu < 0 \) or \( \Delta \mu > 1 \), the TMR will be finite, and vice versa. As shown in Figure 1, the TMR tends to infinity at a certain thickness which will be called “critical thickness” in the following. This means that the TMR is extremely sensitive to a variation in barrier thickness when the barrier thickness is slightly less than the critical thickness. In physics, that is because the critical thickness just corresponds to the critical equation \( \nu(K_h) = \Delta - \mu \), as can been seen in Figure 1.

Let us now turn to the pressure effect. According to ref 8, when \( N \geq 5 \), the band gap of black phosphorus will decrease monotonically with pressure in the range of 0 GPa to 1 GPa, and the change is a few tens of meV/GPa. Considering the case that when the MTJs are without pressure, \( \nu(K_h) \) is slightly larger than \( \Delta - \mu \), and then a pressure is applied to the MTJs. In such a case, \( \nu(K_h) \) will decrease with the pressure and further tend to \( \Delta - \mu \). Accordingly, the TMR will be extremely sensitive to the pressure, and the principle is essentially the same as the thickness effect. This provides a possible working mechanism for the highly sensitive pressure sensors based on MTJs with a black phosphorus barrier, which is illustrated schematically in Figure 2.

In the following, we shall calculate comprehensively the thickness effect and pressure effect of the MTJs with a black phosphorus barrier (the theoretical model used for the calculations and the definition of TMR can be found in the Supporting Information). According to the calculations, we designed in detail the device prototype of the highly sensitive pressure sensors and discuss the advantages and benefits of it.

\section*{RESULTS AND DISCUSSION}

The parameters in the calculations are set as follows. As stated in Theoretical Background, the band gap \( E_g \) of black phosphorus is highly dependent on the thickness. Until now,
there has been no unified expression of the dependence. Here we adopt the expression of ref 5:

$$E_g = A \exp\left(-BN/C\right) + D$$

(2)

where $A = 1.71$ eV, $B = 0.17$, $C = 0.73$, and $D = 0.40$ eV. Because $v(K_h)$ should be approximately proportional to the band gap, we assume that the thickness dependence of $v(K_h)$ is the same as $E_g$. Considering that the thickness of monolayer black phosphorus is 0.524 nm, we shall set

$$v(K_h) = 3.42 \exp\left(-0.17d/0.524 \text{ nm}\right)/(d/0.524 \text{ nm})^{0.73} + 0.8 \text{ eV}$$

(3)

where $d$ is the thickness of the black phosphorus barrier. As for the pressure effect, according to ref 8, the parameters in eq 1 are set as follows: $a = 0.18$ eV/GPa, $\gamma_0 = 1.76$ eV, $P_{coh} = 1.4$ GPa. In addition, because the magnitude of the intralayer primitive translational vector $c = 4.3763 \text{ Å}$, the magnitude of the intralayer reciprocal lattice vector $K_h = 2\pi/c = 1.436 \times 10^{10} \text{ m}^{-1}$.

**Thickness Effect of the TMR in MTJs with a Black Phosphorus Barrier.** First, we investigated the thickness dependence of conductances and TMR under different bias voltages. The theoretical results are depicted in Figure 3 where the chemical potential $\mu$ is 3 eV, half of the exchange splitting $\Delta$ is 5 eV, and the bias $V_0$ is set sequentially as 0, 0.1, 0.5, and 1 V. As shown in Figure 3a, $G_{fp}$ varies nonmonotonically with the barrier thickness, which originates from the interference among the diffracted waves, as pointed out in ref 13. Unlike $G_{fp}$, $G_{AP}$ first decreases with the barrier thickness and tends to zero at a certain thickness, as depicted in Figure 3b. It can be explained as follows: according to eq 3, $v(K_h)$ decreases monotonously with barrier thickness. Physically, the smaller the $v(K_h)$, the less energy the tunneling electrons will acquire. Therefore, the decreasing $v(K_h)$ reduces the number of tunneling electrons having sufficient energy to transit from the spin-up band into the spin-down band. For the case of zero bias limit, as the barrier thickness increases, there will be a critical thickness ($d \approx 1.26$ nm) that corresponds to $v(K_h) = \Delta - \mu$. At this critical thickness, $G_{AP}$ tends to zero because the energy $v(K_h)$ will be insufficient for the incident spin-up electrons to transit into the spin-down band. For the finite bias case, the larger the bias voltage, the greater the critical thickness. This is because the critical thickness corresponds to $v(K_h) = \Delta - \mu - eV_0$ now, which is illustrated schematically in Figure 4. From Figure 4, it can be seen that larger bias can reduce the energy required for the spin-up electrons to transit into the spin-down band, i.e., smaller $v(K_h)$ is needed. According to eq 3, the critical thickness will be greater. Consequently, the TMR will achieve a transition from a finite value to infinity around those critical thicknesses, as shown in Figure 3c. This is just the critical thickness phenomenon presented in Figure 1.

Second, we shall study the thickness dependence of conductances and TMR under different half of the exchange splittings. The theoretical results are depicted in Figure 5 where the chemical potential $\mu$ is 3 eV, the bias is 0.1 V, and the half of the exchange splitting $\Delta$ is set sequentially as 4, 5, and 6 eV. As can be seen in Figure 5b, the critical thickness decreases with the half of the exchange splitting $\Delta$. This can be easily understood: according to the critical equation $v(K_h) = \Delta$.
respectively, for there are critical points located around 1.43 and 1 V, 2, 4, and 8. As shown in Figure 6a, when layers of the black phosphorus barrier depicted in Figure 6 where the chemical potential the black phosphorus barrier. The theoretical results are half of the exchange splitting $\Delta$.

The channel of $T_{11\uparrow}$ on the left side of the critical points, $p^\downarrow$ will be imaginary in some integral regions, which will damage the oscillation property of $\cos[(p^\uparrow - p^\downarrow)d]$. More importantly, it can be seen from Figure 6c that, for $N = 4$ and $N = 8$, TMR decreases from a finite value to infinity around $V_0 = 0.57$ V and $V_0 = 1$ V, respectively. The above two values of $V_0$ meet the critical equation $\nu(K_0) = \Delta - \mu - eV_0$, and this is just the critical bias phenomenon mentioned above. For the cases of $N = 1$ and $N = 2$, $\Delta - \mu - eV_0$ is always smaller than $\nu(K_0)$, so the critical bias phenomenon will not happen. In other words, the critical phenomenon only can happen when critical equation $\nu(K_0) = \Delta - \mu - eV_0$ is met in the variable range.

Pressure Effect of the TMR in MTJs with Black Phosphorus Barrier. As examples, we calculate, respectively, the pressure effect of the TMR for three different thicknesses of the black phosphorus barrier, i.e., $N = 5$, $N = 10$, and $N = 20$. The parameters are chosen to satisfy the condition that, when the MTJs are without pressure, $\nu(K_0)$ is slightly larger than $\Delta - \mu - eV_0$. For the case of $N = 5$, $\mu = 3$ eV, $\Delta = 4.2$ eV, and $eV_0 = 70$ meV, for the case of $N = 10$, $\mu = 3$ eV, $\Delta = 3.8$ eV, and $eV_0 = 50$ meV, and for the case of $N = 20$, $\mu = 3$ eV, $\Delta = 3.7$ eV, and $eV_0 = 70$ meV. The theoretical results are depicted in Figure 7 where the vertical is exponential type. As shown in Figure 7, for all three cases, the TMR increases with the pressure even more rapidly than the exponential form. This is because, according to eq 1, the band gaps of black phosphorus for these three thicknesses all decrease monotonically with pressure within the range 0–1 GPa. In this sense, the monotonically decreasing range of the pressure–band gap curve should be the theoretical range of detection. From eq 1, it can be derived that the greater the number of layers, the

Figure 5. (a) $G_\uparrow$, (b) $G_\downarrow$, and (c) TMR as functions of barrier thicknesses under different half of the exchange splittings $\Delta = 4$, 5, and 6 eV. Here the dashed lines denote the critical barrier thicknesses.

Figure 6. (a) $G_\uparrow$, (b) $G_\downarrow$, and (c) TMR as functions of bias voltage under different numbers of layers of the black phosphorus barrier $N = 1, 2, 4,$ and 8. Here the dashed lines denote the critical bias voltage.
larger the theoretical range of detection: for the number of layers \( N = 5 \), the theoretical range of detection is about 0–1.8 GPa; for \( N = 10 \), the theoretical range of detection is about 0–2.5 GPa; for \( N = 20 \), the theoretical range of detection is about 0–2.8 GPa. It is worth noting that the range of the experimental data in ref 8 only includes 0–2.5 GPa, and therefore the theoretical range of detection for \( N = 20 \) is only theoretically derived from eq 1. In the present case, the sensitivity can be defined as \((\Delta \text{TMR}/\text{TMR}_0)/\Delta P\), where \(\Delta \text{TMR}\) is the change in TMR, \(\text{TMR}_0\) is the TMR under no pressure, and \(\Delta P\) is the change in pressure. It can be deduced from Figure 7 that the highest sensitivity can reach \(8.53 \times 10^5\) MPa\(^{-1}\) at 1 GPa for the case of \(N = 20\). Such high sensitivity originates from the TMR transition at the critical point, as displayed in Figure 2. In principle, the present pressure sensors can reach arbitrary high sensitivity if the measuring range can be sacrificed. This is because, at the critical point of the TMR transition, the TMR has an infinite derivative as well as sensitivity. In theory, we can let \(\Delta = \mu - eV_0\) tend to \(\nu(K_{\text{d}})\) infinitely, which means that the sensitivity can be arbitrarily high. The above results declare that the MTJs with a black phosphorus barrier indeed can be a potential system as highly sensitive pressure sensors.

**Design of Highly Sensitive Pressure Sensors.** Pressure sensors have a wide range of applications, such as transportation, automobile industries, and the medical field. They constitute an important component of the field of micro-electromechanical systems (MEMS) and have the highest market share among all kinds of MEMS sensors. The performance of pressure sensors can be assessed from many aspects; nevertheless, there is no doubt that the sensitivity is a vital parameter. In particular, pressure sensors with high sensitivity are urgently needed in nanotechnology. When the sensitivity is high, good anti-interference, high spatial resolution, and fast response speed are usually necessary. Therefore, the requirements of high sensitivity, good anti-interference, high spatial resolution, and fast response speed are goals for the development of pressure sensors. Unfortunately, conventional types of pressure sensors (e.g., piezoresistive-type sensors and capacitance-type sensors) cannot completely meet the above requirements. This suggests that novel sensing technology should be proposed to improve the corresponding performances.

On the other hand, the applications of MTJs are mainly focused on magnetic memory storage cells and magnetic field sensors. Besides the above familiar applications, M. Löhndorfa et al. developed a type of pressure sensor based on MTJs. Its working mechanism utilizes the inverse magnetostrictive effect (Villary effect), i.e., the shear pressure leads to a change in the magnetization direction of the free layer. In addition, the pressure sensitivity is described by the gauge factor: \(\text{GF} = (\Delta R/R)/\Delta \varepsilon\), where \(\Delta R/R\) is the relative change in resistance and \(\Delta \varepsilon\) is the relative change in strain. Subsequently, a certain amount of research effort has been devoted to improve the performances, especially the GF, of such pressure sensors. These works have initiated a new research direction for both MTJs and pressure sensors and indicate that the pressure sensors based on MTJs have great potential to satisfy the requirements mentioned above. However, owing to the working mechanism, this type of pressure sensor can only measure the shear pressure and is not applicable to the measurement of normal pressure. That confines the applications to certain fields.

Therefore, a new kind of MTJ-based pressure sensor which can measure the normal pressure is needed. The present calculations of the pressure effect demonstrate that the MTJ with a black phosphorus barrier is a good candidate to satisfy this need. Different from previous MTJ-based pressure sensors, the working mechanism now is the TMR transition due to the pressure effect of black phosphorus. According to ref 8, the pressure-induced variation of band gap derives from the decrease of the interlayer distance. In other words, the pressure in the normal direction leads to the variation of TMR. Therefore, the pressure sensors based on MTJs with a black phosphorus barrier are suitable for measuring the normal pressure. To facilitate the practical application, a detailed structure of the device was designed, which is displayed in Figure 8. It can be seen from Figure 8 that two MTJs with different magnetization configurations are in parallel connection to the output parallel current and antiparallel current, respectively. Such a design has two advantages: (1) the parallel current \(I_{\parallel}\) and antiparallel current \(I_{\bar{\parallel}}\) can be simultaneously obtained, which can improve the response speed. (2) If noise exists, it can only influence the magnitudes of \(I_{\parallel}\) and \(I_{\bar{\parallel}}\) but has no influence on the magnitude of TMR. In other words, the present pressure sensors have good anti-interference. In addition, nanometer scale MTJs can lead to miniaturized precision sensors with high spatial resolution. Including the high sensitivity mentioned above, the present pressure sensors possess four advantages and benefits: (1) high sensitivity, (2) good anti-interference, (3) high spatial resolution, and (4) fast response speed. Furthermore, the completely new working
mechanism may lead to a novel research area for both the pressure sensors and the MTJs.

**CONCLUSIONS**

In this paper, we have studied the thickness effect and pressure effect of the MTJs with a black phosphorus barrier. We found a TMR transition phenomenon: the TMR will transition from a finite value to infinity around the critical thickness. The critical thickness can be regulated by both the bias voltage and half of the exchange splitting of electrodes. For the range of numbers of the black phosphorus layers, $5 \leq N \leq 20$, the TMR increases with the pressure more rapidly than the exponential form under appropriate parameters, and the sensitivity can be as high as $8.53 \times 10^3$ MPa$^{-1}$ at 1 GPa for the case of $N = 20$. Such high sensitivity originates from the TMR transition at the critical point. Furthermore, according to the calculations of the pressure effect, we designed in detail the structure of the highly sensitive pressure sensors, and the design diagram is given. The advantages and benefits of the present pressure sensors were discussed. Finally, the present work should not be confined to MTJs with a black phosphorus barrier. It can be extended to MTJs with other band-tunable barriers, such as MoS$_2$.

**ASSOCIATED CONTENT**

*Supporting Information*

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c00748.

Theoretical model for magnetic tunnel junctions with a single-crystal barrier (PDF)

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**Notes**

The authors declare no competing financial interest.

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