Pressure Control of Interface Ferromagnetism in van der Waals Heterostructures

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2. Abstract

$NbSe_2$ and V_5Se_8 are both van der Waals layered materials. By stacking these two materials, it is possible to fabricate an atomically abrupt heterointerface, that is called a van der Waals heterostructure. The Curie temperature of V_5Se_8 was found to increase from 25 K to 40 K by formation of the heterointerface with $NbSe_2$ owing to a unique spin-orbit interaction in $NbSe_2$. However, the detailed mechanism is still in debate. In this study, we examined how the interface effect is modulated by applying external pressure to the $NbSe_2/V_5Se_8$ heterostructure sample. We found that the Curie temperature increases by applying pressure up to 8 GPa, and proved that the interface ferromagnetism can be controlled by the external pressure.

3. Introduction

Ultra-thin van der Waals (vdW) crystals, such as graphene, have been attracting wide attention for the investigation of high-quality two-dimensional (2D) physical properties. Various physical properties such as 2D superconductivity or 2D magnetism have been discovered in ultra-thin vdW materials, but most of the research has focused on the properties of every single substance so far. On the other hand, vdW heterostructures, where different vdW materials are stacked, are recently proposed as the further development way to explore and create unique 2D physical properties as represented by the superconductivity in the twisted bilayer graphene[1,2].

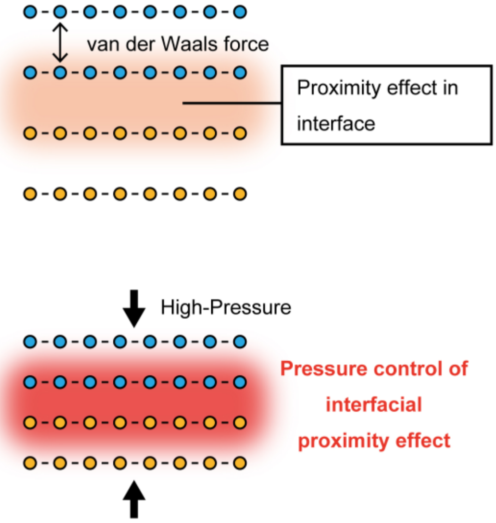

Fig. 1 ：Schematic diagram of this study

In this study, we focus on the pressure effect in a vdW heterostructure. The distance between layers of the vdW crystal is easy to be reduced with respect to isotropic pressure due to its unique highly quasi-2D crystal structures[3,4]. From this background, the proposal of this study is pressure control of the proximity effect in vdW heterointerfaces (Fig. 1). In addition, by systematically measuring the physical properties of the heterostructure with an external control parameter, it is expected that the microscopic information of the interface properties is obtained. There is almost no precedent for the research on the pressure effect in such a vdW heterostructure, except for the recently reported pressure-induced superconductivity in twisted bilayer graphene[5]. Further research will play an essential role to expand the possibilities for the control of physical properties of the van der Waals interfaces.

The target system in this study is the vdW heterostructures of V_5Se_8 and $NbSe_2$. V_5Se_8 is an itinerant ferromagnet[6] and $NbSe_2$ is a non-magnetic metal. $NbSe_2$ has a unique spin-orbit interaction (SOI) called as Zeeman-type SOI due to the breaking of the inversion symmetry in monolayer limit[7] or even the surface of bulk[8]. In the heterostructure of these two materials, the perpendicular magnetic anisotropy is induced in originally isotropic ferromagnetism of V_5Se_8 due to coupling between the giant effective magnetic field of the Zeeman-type SOI and the ferromagnetism. In addition, the Curie temperature, T_C, increases from about 15 K to 40 K. In this study, we investigated how the Curie temperature changes as a result of pressure modulation of the interface effect.

4. Experiment

4.1. Thin film fabrication

VdW heterostructures of ferromagnetic V_5Se_8 and metallic $NbSe_2$ were fabricated on sapphire substrates by molecular beam epitaxy. In the fabrication process, sapphire substrates were annealed at a high temperature and surface-treated, followed by the supply of transition metals V and Nb using an electron beam evaporator to grow heterostructures of sapphire/V_5Se_8/$NbSe_2$. Selenium was supplied through the whole process. Since ultra-thin $NbSe_2$ is easily oxidized and unstable in air, we deposited selenium with ~100 nm thickness at room temperature to form a cap layer after the growth.

4.2. Evaluation of transport properties

The transport properties of each fabricated sample, such as the anomalous Hall effect and the longitudinal resistance, were evaluated. As shown in Fig. 2, the temperature at which the signal of the anomalous Hall effect arises corresponds to the temperature at which the resistance anomaly appears, showing T_C is around 35 K. The anomaly in the temperature dependence of the resistance indicates that the electron scattering is suppressed by the ferromagnetism. Now it is confirmed that the Curie temperature is estimated only by measuring the temperature dependence of the resistance.

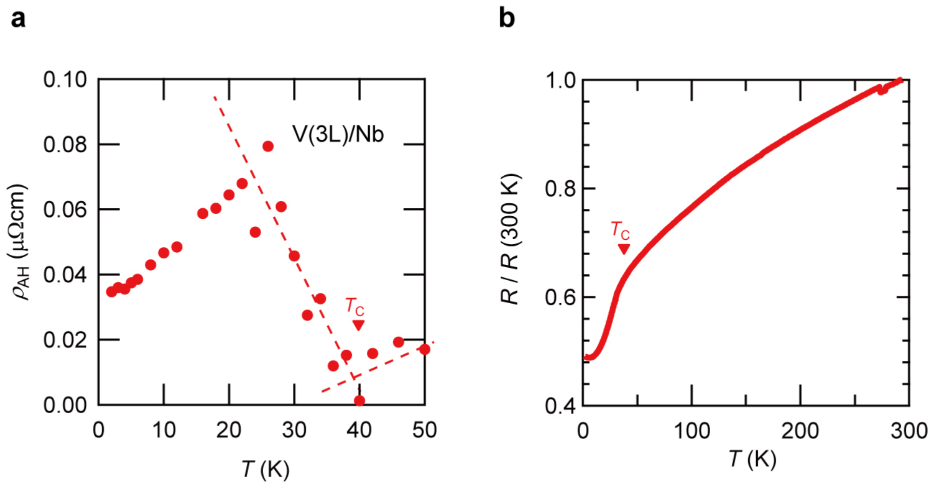

Fig. 2： Temperature dependence of the amplitude of the anomalous Hall effect (a) and the longitudinal resistance (b) in V_5Se_8 (3 L)/$NbSe_2$ (4 L). The arrow indicates the T_C ~ 35 K.

4.3. Cap layer

In order to measure the electrical transport properties by the four-terminal method under pressure, it is necessary to firmly bond the gold wire to the sample. Therefore, thermosetting conductive adhesives are often used. To cure the commonly used conductive adhesive, the sample needs to be heated at 120 ℃ for about 30 minutes. After this heating, the anomaly disappeared from the temperature dependence of the resistance as shown in Fig. 3. This is probably because the new conductive layer was formed by the chemical reaction between the Se cap layer and the adhesive. Therefore, we tried to cap the sample with amorphous Al_2O_3 to avoid the chemical reaction. To deposit Al_2O_3 cap layer, we used Kawasaki lab's ALD (Atomic Layer Deposition) equipment. The thickness of Al_2O_3 cap layer is 30 nm. After depositing Al_2O_3 layer, the electrical resistance has the anomaly even after heated to cure the adhesive.

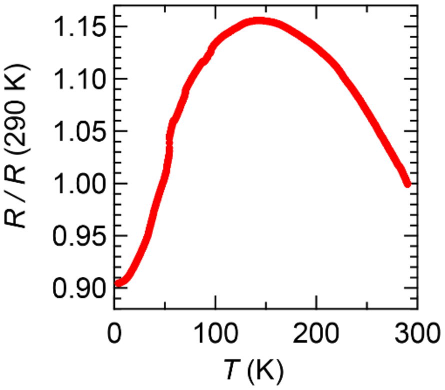

Fig. 3 ：Temperature dependence of the resistance after heating. (Without Al_2O_3 cap)

4.4. Application of pressure

This study is performed to evaluate the electrical transport properties of thin films under high pressure. Therefore, a cubic anvil high-pressure apparatus was used in this study because the method for measuring electrical conductivity was established. We have used the constant-loading cubic anvil high-pressure apparatus at Uwatoko lab., ISSP. The cubic anvil high-pressure apparatus can generate high pressure up to 10 GPa. The pressure range is much wider compared to 0-3 GPa of a piston-cylinder cell which is often used in high-pressure experiments. In addition, since the measurable sample size is relatively large(～1 mm), electrical conductivity measurements can be stably performed.

Figure 4a is a diagram that shows how the cubic anvil high-pressure apparatus generate high pressure. Ultra-high hydrostatic pressure is generated by pressing a cubic gasket isotropically from six directions using anvils. The sample is sealed inside the Teflon cell as shown in Fig. 4b, and the Teflon cell is filled with glycerin as a pressure medium. The anvils make electrical contacts to the four terminals of the sample from the four sides of the gasket.

The $V_5Se_8/NbSe_2$ heterostructure thin film on a sapphire substrate was cut out in a size of about 0.6 mm × 0.4 mm × 1.0 mm and four terminals were attached using a conductive adhesive as shown in Fig. 4c. The electrical resistance was measured under the maximum pressure of 8 GPa, and the pressure dependence of the Curie temperature was evaluated from the anomaly observed in the temperature dependence of the electrical resistance.

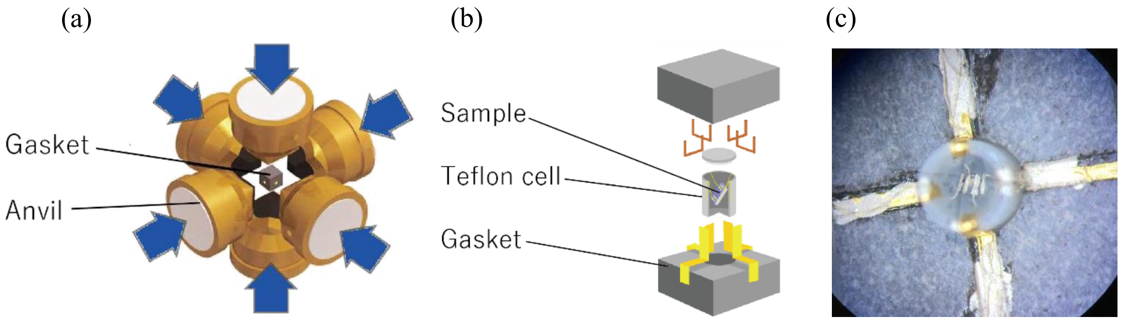

Fig. 4：Schematic diagram of the cubic anvil apparatus(a), Sample space inside the Teflon cell (b), The photo of the sample (c).

5. Result & Discussion

Figure 5a shows the temperature dependence of the resistance under pressure of a thin film having V_5Se_8 (3 L) / $NbSe_2$ (4 L) heterostructure. The temperature at which the anomaly appears in the resistance at ambient pressure is $T_C = 37$ K. It corresponds to the Curie temperature obtained from the temperature dependence of the anomalous Hall effect.

The temperature at which the anomaly appears in the resistance increases as applied pressure increases. The Curie temperature is $T_C = 37$ K at $P = 0$ GPa and it reaches around $T_C = 51$ K at $P = 8$ GPa. dT_C/dP is about +3.5 (K/GPa) around $P = 0$ GPa. Figure 5b shows the pressure dependence of the Curie temperature.

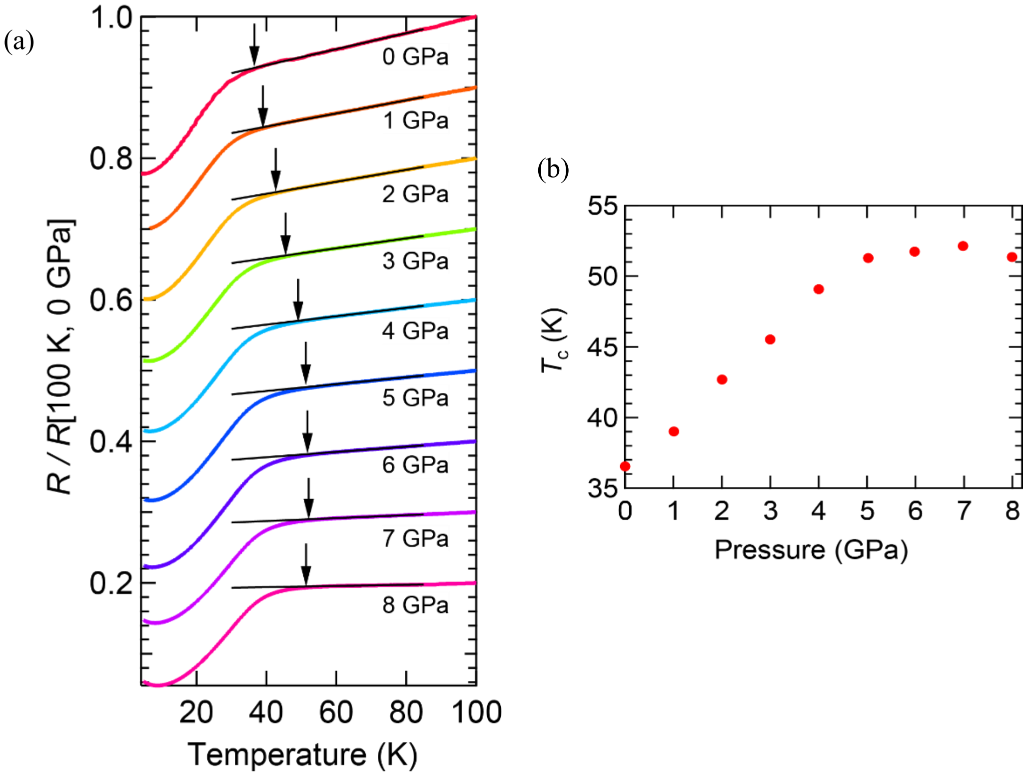

Fig. 5：Temperature dependence of the resistance under pressure (a), pressure dependence of the Curie temperature T_C (b). The arrows in (a) indicate the T_C. For clarity, the curves are normalized by the resistance at 0 GPa, 100 K and successively shifted.

V_5Se_8 is an itinerant magnet (Néel temperature T_N = 34 K, Curie temperature T_C = 15 K), however, the increase of the Curie temperature under pressure in the V_5Se_8 / $NbSe_2$ heterostructure is in contrast to the decrease of the Curie temperature in typical itinerant ferromagnets such as an Fe-Co-Ni alloy system[9,10]. In the itinerant band ferromagnets, the value of the ferromagnetic moment is determined by the competition between the Coulomb repulsion energy and the kinetic energy (bandwidth). When external pressure is applied, the band width increases as the interatomic distance decreases, and the number of electrons at the Fermi level decreases. Therefore, it is expected that the ferromagnetic transition temperature decreases when pressure is applied. This experimental result cannot be explained just because of the typical picture that V_5Se_8 is an itinerant band ferromagnet. The possible mechanisms are discussed as follows:

(i) The response from V_5Se_8 with the localized magnetic moments and conductive layers.

According to the picture that V_5Se_8 is a typical band ferromagnet, the experimental result with the enhancement of T_C under the high-pressure is contrary to the response expected from single-V_5Se_8.

However, one possible explanation considering without the heterointerface is that V_5Se_8 is not a simple band ferromagnet but the itinerant ferromagnet where the localized magnetic moment and conductive layers coexist. V_5S_8 has the same crystal structure as V_5Se_8 where the V sites are separated into two types: the atoms with the same crystal structure as $1T$-VS_2 and the intercalated atoms into inter-layers. It is confirmed that the magnetic moments of V_5S_8 nearly localize around the intercalated atoms[11]. Since the pressure response is considered to depend on the exchange interaction between conductive layers and the localized moments, it cannot be concluded that the Curie temperature decreases.

(ii) The modulation of the interface proximity effect.

The last scenario is that the interface effect of the V_5Se_8 / $NbSe_2$ vdW heterointerface, which originally increased the Curie temperature, was enhanced by the application of pressure. The interface proximity effect in V_5Se_8 / $NbSe_2$ that increases T_C and induces the perpendicular magnetic anisotropy could be either of following two types of mechanism: [1] the exchange interaction between ferromagnetic V_5Se_8 and the magnetic moments induced in $NbSe_2$ and [2] Rudermann-Kittel-Kasuya-Yoshida (RKKY) interaction with the localized magnetic moments in V_5Se_8 and conduction electrons in $NbSe_2$. In both scenarios, the value of the *s-d* exchange interaction $J_{\text{V-Nb}}$ between the magnetic moments of V_5Se_8 and conducting $NbSe_2$ contributes to the enhancement of Curie temperature. Therefore, it is possible that the Curie temperature has increased due to the increase in $J_{\text{V-Nb}}$ with the application of pressure.

6. Summary & Future works

In this self-directed joint research, the pressure was applied to the ferromagnetic thin film of V_5Se_8 / $NbSe_2$ heterostructure, and the Curie temperature increased with increasing pressure. To confirm that this result is originated from the modulation of interface effect in the vdW heterostructure, it is necessary that the resistance of the heterostructure of single-layer V_5Se_8 and $NbSe_2$ is measured under pressure. In addition, the magnetic transition is currently evaluated only by the temperature dependence of the resistance under the zero magnetic field. By measuring magnetoresistance or anomalous Hall effect under pressure up to several GPa, it is expected that further discussions can be held.

7. Acknowledgement

We are grateful to our supervisors Prof. T. Shibauchi and Prof. Y. Iwasa for their support and cooperation in carrying out this research. We also thank Prof. Y. Uwatoko for his great support and cooperation in the resistance measurements under pressure. We also thank Prof. M. Nakano for his great support and cooperation in the film growth. We would also like to thank Prof. K. Kanoda and Prof. Y. Nakamura, assistant supervisors of the MERIT program, for their willingness to approve the proposal for this research. We would like to thank the MERIT program for giving us this valuable opportunity.

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