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ABSTRACT

Magnetism in two-dimensional van der Waals materials has received significant attention recently. The Curie temperature reported for these materials, however, has so far remained relatively low. Here, we measure magneto-optical Kerr effects under a perpendicular magnetic field for van der Waals ferromagnet Cr$_2$Ge$_2$Te$_6$ and its heterostructures with antiferromagnetic insulator NiO. We observe a notable increase in both Curie temperature and magnetic perpendicular anisotropy in Cr$_2$Ge$_2$Te$_6$/NiO heterostructures compared to those in Cr$_2$Ge$_2$Te$_6$. Measurements on the same exfoliated Cr$_2$Ge$_2$Te$_6$ flake (on a SiO$_2$/Si substrate) before and after depositing NiO show that the hysteresis loop can change into a square shape with a larger coercive field for Cr$_2$Ge$_2$Te$_6$/NiO. The maximum Curie temperature ($T_C$) observed for Cr$_2$Ge$_2$Te$_6$/NiO reaches $120 \text{ K}$, which is nearly twice the maximum $T_C \approx 60 \text{ K}$ reported for Cr$_2$Ge$_2$Te$_6$ alone. Both enhanced perpendicular anisotropy and increased Curie temperature are observed for Cr$_2$Ge$_2$Te$_6$ flakes with a variety of thicknesses ranging from $5 \text{ nm}$ to $200 \text{ nm}$. The results indicate that magnetic properties of two-dimensional van der Waals magnets can be engineered and controlled by using the heterostructure interface with other materials.

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Recently, magnetism in layered van der Waals (vdW) materials has attracted great attention because of their unique magnetic properties stemming from their two-dimensional (2D) nature. Such layered vdW materials provide an opportunity to fabricate heterostructures free from constraints in conventional film growth and promise a unique route to explore new functionality of these materials based on the electric field and crystalline symmetry. One of the major issues in vdW ferromagnets is that the ferromagnetic transition temperature (Curie temperature) is relatively low. Cr$_I_2$ and Cr$_2$Ge$_2$Te$_6$ were reported as atomically thin ferromagnets in 2017, where the Curie temperature ranges from 30 K (bilayer Cr$_2$Ge$_2$Te$_6$) to 45 K (monolayer Cr$_I_2$), being intriguingly low compared to the bulk values of 61 K (bulk Cr$_I_2$) and 66 K (bulk Cr$_2$Ge$_2$Te$_6$). To enhance the Curie temperature, a variety of approaches using interface and gap engineering have been proposed including the dielectric effect, spin–orbit coupling proximity, charge transfer, and interface hybridization. However, only a few approaches have been implemented and reported so far, such as electric gating. Therefore, it is important to search for other effective approaches to enhance the Curie temperature. Here, we study magnetic properties in heterostructures between antiferromagnet NiO and vdW ferromagnet Cr$_2$Ge$_2$Te$_6$. We will report magneto-optical Kerr effects (MOKEs) and detect hysteresis arising from ferromagnetism.

Cr$_2$Ge$_2$Te$_6$ was earlier reported by Carteaux et al., and recently, there is significantly increasing interest because this is a layered two-dimensional magnet with mechanical cleavability down to atomically thin layers. The Cr$_2$Ge$_2$Te$_6$ single crystals used in this work were grown via a self-flux technique. First, 100 mg of Cr powder, 200 mg of Ge powder, and 2 g of Te were sealed in a quartz tube. The mixture
was heated to 1050 °C and held for 30 h, and then, it was cooled down to 475 °C in 10 days, and finally, the Ge-Te flux was removed using a centrifuge at this temperature. The magnetic properties of the bulk crystals were characterized by magnetometry using the MPMS (magnetic property measurement system), and the Curie temperature was found to be $T_C \approx 66$ K from the minimum of $dM/dT$ curves (measured with the magnetic field of 50 mT in the $c$-axis). The observed magnetic properties were consistent with the previous reports.\(^1\),\(^4\),\(^7\)–\(^13\) Cr$_2$Ge$_2$Te$_6$ crystals were mechanically cleaved onto a silicon substrate in ambient conditions (with the SiO$_2$ thickness of 285 nm). The thickness of the flakes was characterized by using an atomic force microscope. Atomically thin flakes were visible with thickness-dependent color contrast due to the interference effect as shown in Fig. 1(a).

Polar magneto-optical Kerr Effect (MOKE) measurements were performed in an Oxford MicrostatMO system in the Faraday configuration. A temperature stabilized laser diode (635 nm in wavelength) was used to deliver a linearly polarized laser beam that was focused onto the sample at normal incidence using a 0.6 NA 100X long working distance objective. The estimated power delivered to the sample is less than 3 $\mu$W. The laser beam was intensity modulated using a chopper, and the reflected beam was sent through a Wollaston prism oriented at 45° with respect to the initial polarization to split the beam into two. The split beams were collected by two photodiodes arranged in a differential mode whose output was sent to an SR570 current pre-amplifier (Stanford research systems). The output of the amplifier was then measured using a lock-in referenced to the chopper frequency. In order to complete the measurements, each photodiode was covered in turn to obtain the total intensity signal. This is used to normalize the previously measured signals that will yield a quantity proportional to the Kerr effect signal (rotation of the polarization angle). Calibration was performed to obtain the actual conversion factor between radians and the normalized signal by tilting the polarizer by a few fractions of a degree and recording the normalized signal.

Figure 1(c) shows MOKE curves (Kerr rotation angle vs magnetic field) for a sample before NiO deposition. The magnetic field was applied perpendicular to the substrate because the easy axis of Cr$_2$Ge$_2$Te$_6$ was reported in that direction. For the flakes with the thicknesses more than 5 nm, we observed clear hysteresis. For the flakes with a thickness of 5 nm or less, we did not observe clear hysteresis, which can be attributed to the weaker two-dimensional magnetism\(^1\) and/or to the possibly not-pristine surface or film degradation (e.g., oxidization). After a 20-nm-thick NiO layer was deposited, we measured the same flakes for comparison. These flakes, for the ones that showed a clear hysteresis before the deposition, showed an increase in the coercive field and a change in the hysteresis into a rectangular shape as shown in Fig. 1(d). This clearly indicates the enhanced perpendicular anisotropy induced by depositing the NiO layer.

Importantly, Cr$_2$Ge$_2$Te$_6$/NiO not only enhances the perpendicular anisotropy but also increases the Curie temperature. Figure 2(a) shows the temperature dependence of the hysteresis curves in the MOKE signal for the sample shown in Fig. 1 (Cr$_2$Ge$_2$Te$_6$ thickness 7 nm, position 2). We characterized the Curie temperature as the midpoint between the temperature in which the coercive field is no longer seen and the one in which we still see a coercive field. For position 2, we observed that the Curie temperature increased by 30 K to ~85 K after NiO deposition. Similarly, we observed an increased Curie temperature to 85 K at position 1 (Cr$_2$Ge$_2$Te$_6$ thickness 8 nm), to 70 K at position 3 (Cr$_2$Ge$_2$Te$_6$ thickness 6 nm), and to 70 K at position 4 (Cr$_2$Ge$_2$Te$_6$ thickness 5.5 nm). For comparison, we measured another sample with 50 nm thickness of NiO, while the thickness of the flake was similar to that of position 2. We observed an even larger increase in the Curie temperature up to 115 K as shown in Fig. 2(b), which is about twice of the Curie temperature on the Cr$_2$Ge$_2$Te$_6$ flakes without NiO. In this sample, we also observed further enhanced perpendicular anisotropy as the coercive field was even higher than that of a sample with a NiO thickness of 20 nm.

Figure 2(c) shows the hysteresis of a 202-nm-thick Cr$_2$Ge$_2$Te$_6$ flake with NiO of 20 nm. The hysteresis loop at T = 7 K shows small

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**FIG. 1.** (a) and (b) Optical microscopy image of Cr$_2$Ge$_2$Te$_6$ (CGT) flakes on a Si/SiO$_2$ substrate, before (a) and after (b) the deposition of NiO. After the deposition of NiO with a thickness of 20 nm, the optical contrast changes because the interference condition is modified. The scale bar is 10 $\mu$m. (c) Measured magneto-optical Kerr effect (MOKE) curves at the temperature of 7 K with the magnetic field perpendicular to the substrate. The curves from different positions (labeled 1–6) are shifted vertically. The positions are marked in the images (a) and (b). The CGT-flake thickness at each position was measured by atomic force microscopy. (d) MOKE curves measured at the same positions after NiO deposition. Perpendicular anisotropy is strongly enhanced, and square shaped hysteresis curves are observed. The opposite sign of the MOKE hysteresis curves between (c) and (d) is supposedly due to the different optical constants of the sample without (c) and with (d) the NiO overlayer.\(^1\)}
relatively thick Cr$_2$Ge$_2$Te$_6$ that we could be accessing to the (different) such thick flakes. We also raise the possibility in heterostructures of becomes more rectangular in shape and indicates the presence of above 55 K in Fig. 2(c) separately from that of bulk Cr$_2$Ge$_2$Te$_6$ itself measured in bulk Cr$_2$Ge$_2$Te$_6$, se et esupplementary material). At ele-

perpendicular anisotropy (coercive field of nearly 0 Oe, similar to that measured in bulk Cr$_2$Ge$_2$Te$_6$, see the supplementary material). At elevated temperatures (T = 55 K and above, where ferromagnetism dis-

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metal and 2p electrons in oxygen is known to induce high perpendicular anisotropy for MgO/Fe and MgO/Co from both experimental and theoretical studies,\(^1\) which may provide a microscopic mechanism for the enhanced perpendicular anisotropy and increase in the Curie temperature observed in the present studies. We also note that we did not observe any exchange bias (for example reported earlier for the NiO/Ni\(_{81}\)Fe\(_{19}\) interface\(^1\)) in our hysteresis loop. It is noted that the magnetic field was not applied when we made the interface during the NiO deposition.

In summary, we studied the magnetic properties of a two-dimensional van der Waals magnet and Cr\(_2\)Ge\(_2\)Te\(_6\) flakes covered by NiO thin films by magneto-optical Kerr effects. We characterized the hysteresis loops of Cr\(_2\)Ge\(_2\)Te\(_6\) flakes before and after NiO deposition. Cr\(_2\)Ge\(_2\)Te\(_6/\)NiO showed a strong increase in Curie temperature and a clear enhancement in perpendicular anisotropy as evidenced by the increase in the coercive field and the change in hysteresis into a rectangular shape. We observed the Curie temperature as high as 115 K for Cr\(_2\)Ge\(_2\)Te\(_6)/\)NiO with a NiO thickness of 50 nm, more than twice the one for Cr\(_2\)Ge\(_2\)Te\(_6\) without NiO. The Curie temperature increase for Cr\(_2\)Ge\(_2\)Te\(_6/\)NiO is observed for Cr\(_2\)Ge\(_2\)Te\(_6\) with thicknesses ranging from 5 nm to 200 nm. Even Cr\(_2\)Ge\(_2\)Te\(_6/\)NiO with 200 nm thick Cr\(_2\)Ge\(_2\)Te\(_6\) flakes showed the higher Curie temperature than bulk Cr\(_2\)Ge\(_2\)Te\(_6\). These results indicate the magnetic properties of two-dimensional van der Waals materials can be controlled by employing the heterostructure and interface with other materials.

See the supplementary material for the hysteresis curve of bulk Cr\(_2\)Ge\(_2\)Te\(_6\) and XRD data of Si/SiO\(_2/\)Pt/NiO.

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