

Effect of hydrostatic pressure on ferromagnetism in two-dimensional CrI₃Suchanda Mondal,¹ Murugesan Kannan,² Moumita Das,¹ Linganani Govindaraj,² Ratnadwip Singha,¹ Biswarup Satpati,¹ Sonachalam Arumugam,² and Prabhat Mandal¹¹*Saha Institute of Nuclear Physics, HBNI, 1/AF Bidhannagar, Calcutta 700 064, India*²*Centre for High Pressure Research, School of Physics, Bharathidasan University, Tiruchirappalli 620 024, India*

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We have investigated the magnetic properties of the highly anisotropic layered ferromagnetic semiconductor CrI₃ under hydrostatic pressure with the magnetic field along the easy axis of magnetization. At ambient pressure, CrI₃ undergoes a second-order paramagnetic-to-ferromagnetic phase transition at $T_C = 60.4$ K. T_C is found to increase sublinearly from 60.4 to 64.9 K as pressure increases from 0 to 1.0 GPa. With the increase in pressure, the transition becomes sharper while magnetization at low field decreases monotonically due to a decrease in magnetocrystalline anisotropy. The weak low-field anomaly at around 48 K, resulting from two-step magnetic ordering, also shifts toward higher temperatures with increasing pressure. The observed increase in T_C and the decrease in magnetization could originate from a change in coupling between the layers and the Cr-I-Cr bond angle with pressure.

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Two-dimensional ferromagnetic (FM) semiconductors exhibit a wide range of novel electronic properties with an immense potential for applications in different magnetoelectronic technologies [1–4]. In spintronic devices, instead of or in addition to charge degrees of freedom, electron spin is used for information storage and processing [5–9]. This can be realized when magnetism is incorporated into the active materials. Low-dimensional magnetic systems with intrinsic ferromagnetism have also generated considerable interest for fundamental research due to the occurrence of several quantum phenomena [10–15]. According to the Mermin-Wagner theorem, the two-dimensional (2D) isotropic Heisenberg model does not show long-range magnetic ordering at any finite temperature [11]. On the other hand, a 2D Ising system does order at finite temperature, as predicted by Onsager in his remarkable paper [10]. Recently, two different types of layered FM semiconductors with weak interlayer van der Waals interactions, namely, chromium trihalides, CrX₃ ($X = \text{Cl, Br, I}$), and chromium-based ternary compounds, Cr₂Z₂Te₆ ($Z = \text{Si, Ge}$), have received considerable attention as promising candidates for possible applications in spintronic technology owing to their easy exfoliation property and the existence of long-range ferromagnetism across the bilayer or within a monolayer [16–23]. Among different members of Cr₂Z₂Te₆ and CrX₃ series, bulk Cr₂Ge₂Te₆ and CrI₃ compounds exhibit the highest FM transition temperature (T_C), 68 and 61 K, respectively. Although their T_C 's are comparable, in contrast to Cr₂Ge₂Te₆, the magnetic interaction is much more anisotropic in CrI₃ due to the very weak coupling between the layers. The ferromagnetism is retained in a single layer, with a transition temperature as high as 45 K. It is, in fact, very interesting that CrI₃ is antiferromagnetic (AFM) in bilayers [18]. Also, an extremely high tunneling magnetoresistance (10⁵%) could be observed in an exfoliated thin film of CrI₃ [21].

Unlike chemical substitution, hydrostatic pressure (P) is a continuously tunable thermodynamic variable which can be

used to tune the phase transition as well as charge conduction mechanism without introducing any disorder in the system. Primarily, pressure alters the bond lengths and angles of the lattice which, in turn, affect the intersite electron hopping process. As a result, both transport and magnetic properties may change significantly with the application of pressure. For example, several systems undergo a pressure-induced insulator-to-metal transition [24–26]. Even exotic phenomena such as superconductivity at close to room temperature can be achieved by applying high pressure [27,28]. In perovskite manganites, both electronic and magnetic subsystems are found to be highly susceptible to external pressure as well as chemical pressure arising out of a variation in ionic size [29–31]. However, the role of pressure on T_C in an insulating ferromagnet is a very complicated and debatable issue due to the competition between the direct exchange and superexchange. It is believed that the superexchange interaction strength increases with compression, and as a result, the pressure coefficient of T_C increases linearly with isothermal compressibility [32]. Although T_C has been found to increase with an increase in P in several insulating ferromagnets, there are examples where T_C decreases with applied pressure [32]. Very recently, the magnetic properties of Cr₂Ge₂Te₆ have been investigated under pressure and T_C was found to decrease monotonically with pressure up to 1 GPa [33]. Also, a small but negative pressure coefficient of T_C has been reported for CrBr₃ [34]. On the other hand, T_C increases above a critical value of applied pressure in layered VI₃ [35]. So, it is important to investigate systematically the role of pressure on ferromagnetism in several insulating layered ferromagnets. In the present Rapid Communication, we have studied the effect of hydrostatic pressure on the magnetic properties of a well-characterized single-crystalline sample of CrI₃. In contrast to Cr₂Ge₂Te₆ and CrBr₃, T_C in CrI₃ is observed to increase monotonically with pressure. Also, with increasing pressure, the paramagnetic-ferromagnetic (PM-FM) transition becomes

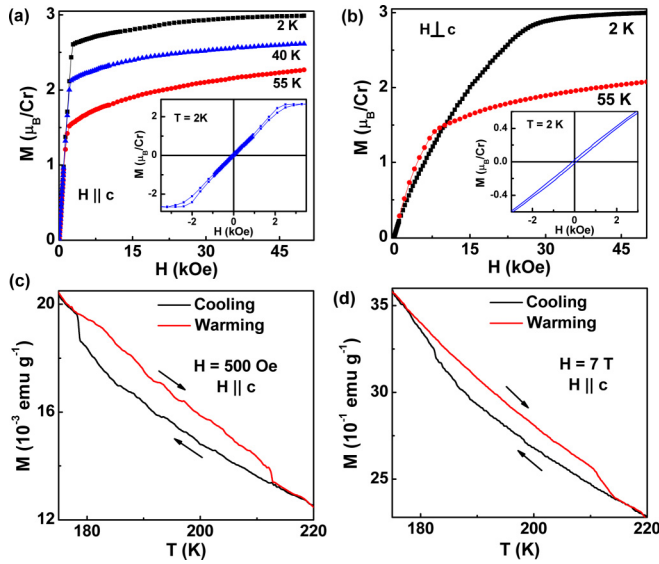


FIG. 1. (a) and (b) Field dependence of magnetization for a CrI_3 crystal measured at different temperatures with applied field parallel to the c axis and perpendicular to the c axis, respectively. The insets show a hysteresis loop in the low-field regime. The observed behavior is qualitatively similar to what has been reported earlier [16,19]. (c) and (d) Field-cooled magnetization for heating and cooling cycles with a magnetic field 500 Oe and 7 T, respectively, applied along the c axis. A clear anomaly along with thermal hysteresis can be seen below ~ 212 K due to the first-order structural transition.

sharper and the value of magnetization with the field parallel to the c axis decreases.

Single crystals of CrI_3 were grown by the standard chemical vapor transport technique using high-purity elements. Analyses of the phase purity and structural details were carried out using both high-resolution x-ray diffraction and transmission electron microscopy. The magnetic measurements were performed in a 7-T superconducting quantum interference device vibrating sample magnetometer (SQUID-VSM). The bulk single crystal of CrI_3 undergoes a long-range ferromagnetic ordering below $T_C \sim 60.4$ K. Details of the sample preparation, characterization, and magnetic measurements are provided in the Supplemental Material [36].

Figures 1(a) and 1(b) show isothermal magnetization curves at a few representative temperatures with the field parallel to the c axis ($H \parallel c$) and perpendicular to the c axis ($H \perp c$), respectively. Well below T_C , magnetization increases sharply and starts to saturate above 3 kOe for $H \parallel c$, whereas M approaches saturation at a much higher field for $H \perp c$, above 30 kOe. This indicates that CrI_3 is a highly anisotropic system and the c axis is the easy axis of magnetization. Both the magnitude and nature of the field dependence of M are very similar to the observations made earlier [16,19]. Figure 1(a) shows that M decreases monotonically with an increase in temperature as in the case of a ferromagnet, but the critical field (H_{sat}) above which M exhibits saturationlike behavior does not change significantly with temperature. However, M exhibits an anomalous behavior for $H \perp c$. With increasing temperature, though, M decreases at high fields, it shows a small increase in the low-field region. Figure 1(b) shows that M is slightly higher for 55 K as compared to that for 2 K when

the applied field is below 1 T. Above 1 T, M increases slowly. Thus, for $H \perp c$, H_{sat} decreases rapidly with increasing temperature. Similar behavior has been observed in the layered itinerant ferromagnet Fe_3GeTe_2 [37]. This unusual behavior of M is due to the strong suppression of magnetocrystalline anisotropy with increasing temperature [38]. The value of the saturation moment at 2 K and 5 T is about $2.98 \mu_B/\text{Cr}^{3+}$ ion, which is very close to the expected moment ($3 \mu_B$) in the high-spin configuration state of the Cr^{3+} ion. The insets of Figs. 1(a) and 1(b) show the hysteresis loop in the low-field region for $H \parallel c$ and $H \perp c$, respectively. In both cases, the hysteresis is very weak. The coercive field is about 72 Oe for $H \parallel c$ and 85 Oe for $H \perp c$, which are close to an earlier report [16]. The weak coercivity suggests that CrI_3 is a soft ferromagnet similar to CrGeTe_3 and CrSiTe_3 [33].

CrI_3 exhibits a first-order structural phase transition from monoclinic to rhombohedral below T_S [16]. In order to investigate how this transition affects magnetic properties, magnetization has been measured by cooling the sample from room temperature down to 2 K in the presence of field and subsequent warming, as shown in Figs. 1(c) and 1(d). M displays an anomaly around $T_S \sim 212$ K, and the cooling and heating cycle data do not overlap with each other but display a significant thermal hysteresis over a wide range, 212–180 K, which is consistent with the reported temperature-dependent x-ray diffraction [16]. Although the value of M changes systematically with H , the nature of the $M(T)$ curve in the range 212–180 K is found to remain insensitive to fields up to 7 T. We have also performed magnetization measurements with $H \perp c$ and observed similar behavior.

To capture the salient features of ferromagnetism under pressure, field-cooled magnetization with $H \parallel c$ has been plotted in Fig. 2(a) as a function of temperature for different applied pressures. With an increase in P , M decreases monotonically and the transition region shifts slowly toward higher temperatures. The increase in T_C with an increase in P suggests that pressure stabilizes the ferromagnetic state. However, the sharp decrease in M with increasing pressure is quite unusual and appears to be inconsistent with this simple picture. Usually, in a simple ferromagnet, the magnetic moment is expected to increase when T_C increases with an application of pressure. In $\text{Cr}_2\text{Ge}_2\text{Te}_6$, M shows a nonmonotonic dependence on P [33]. Initially, M increases with pressure and then decreases slowly. M remains slightly smaller at ambient pressure. In order to determine the dependence of T_C on pressure in CrI_3 , T_C has been estimated from the dM/dT vs T curves. For each pressure, the $dM(T)/dT$ curve shown in the inset of Fig. 2(a) exhibits a very sharp minimum and it shifts progressively toward higher temperatures with increasing pressure. Indeed, we observe that the full width at half minimum of the dM/dT vs T curve is smaller for $P > 0$. In particular, the transition is very sharp for 0.1 GPa. Thus, the PM-FM transition in CrI_3 remains sharp with an application of pressure up to 1 GPa. In $\text{Cr}_2\text{Ge}_2\text{Te}_6$, the transition becomes broader under pressure [33]. The pressure variation of T_C is demonstrated in Fig. 2(b), which shows that the dependence of T_C on pressure is not linear. Initially, T_C increases very rapidly with pressure at a rate of ~ 12 K/GPa up to about 0.10 GPa and then continues to increase further, albeit at a slower rate. For $P \geq 0.5$ GPa, T_C increases approximately linearly

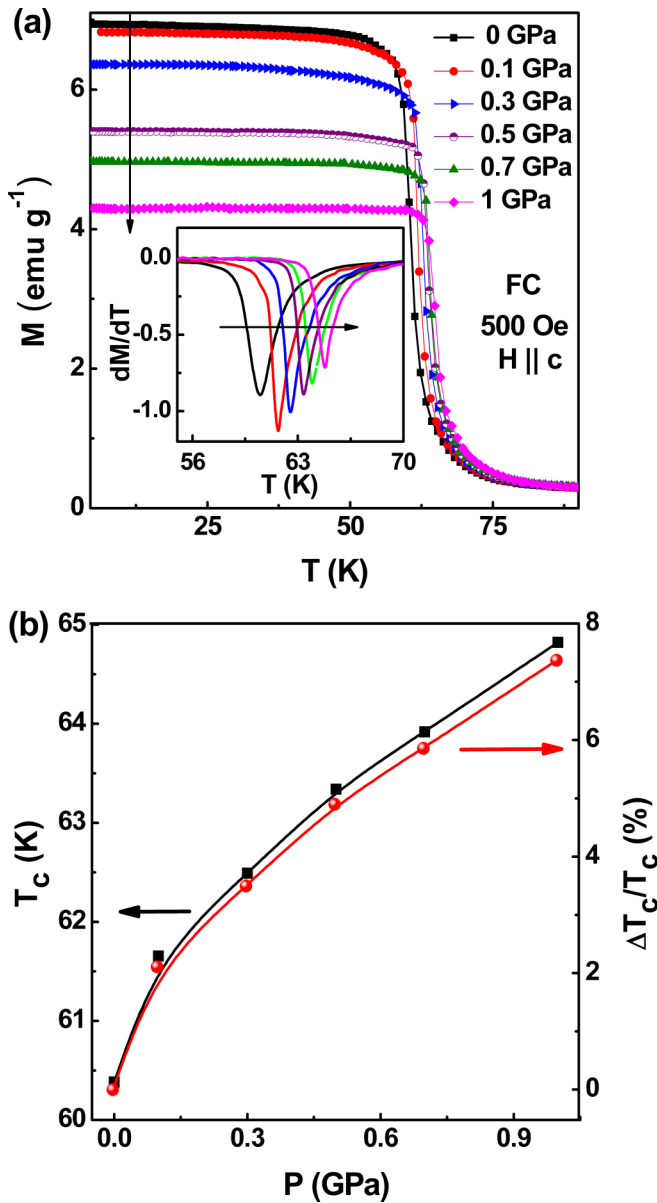


FIG. 2. (a) Temperature dependence of field-cooled magnetization for a CrI_3 crystal at different applied pressures with the magnetic field parallel to the c axis. The inset shows dM/dT as a function of T for different pressures. Arrows indicate the direction of the increase in pressure. (b) T_C , determined from the position of the minimum in dM/dT curves, along with the relative change in T_C ($\Delta T_C/T_C$), are plotted as a function of pressure.

with a pressure coefficient $dT_C/dP \sim 3$ K/GPa. We have also calculated the relative change in T_C , defined as $\Delta T_C/T_C = [T_C(P) - T_C(0)]/T_C(0)$, which has been plotted along with T_C . This plot shows that T_C increases by about 7.5% for an applied pressure of 1 GPa.

In order to understand and explain the systematic decrease of magnetization with applied pressure in Fig. 2(a), we have measured the magnetic field dependence of M below T_C with an $H\parallel c$ configuration. In Fig. 3(a), the magnetic moment under 1 GPa pressure has been plotted as a function of H at a temperature of 5 K. In the same figure, $M(H)$ curves at ambient pressure for the $H\parallel c$ and $H\perp c$ configurations are also

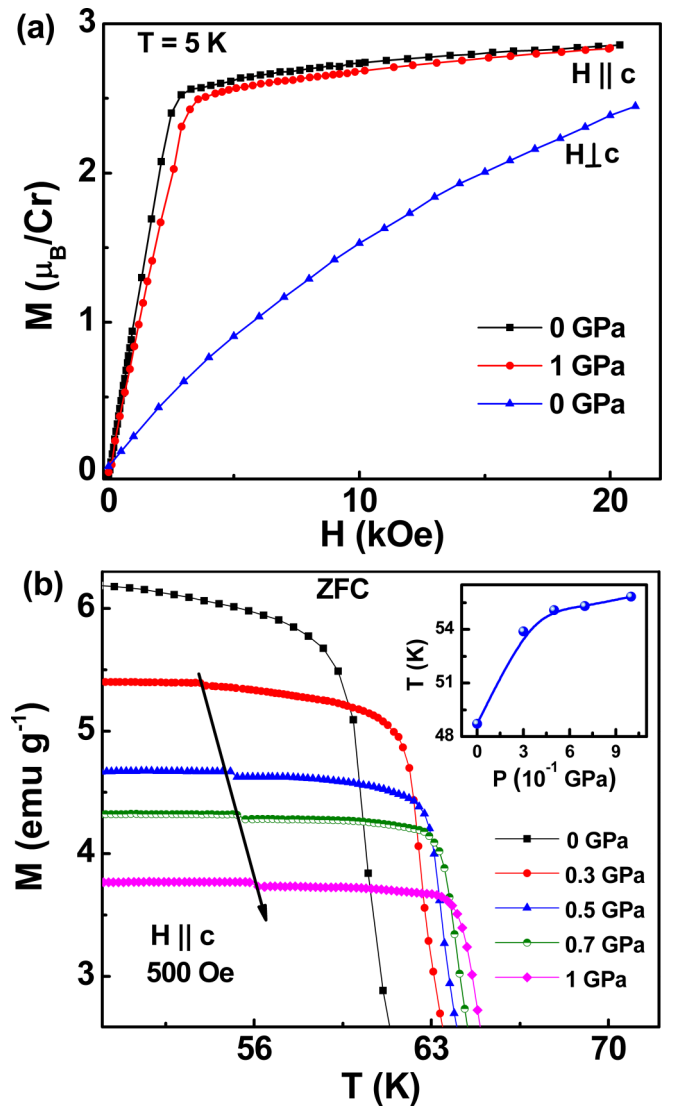


FIG. 3. (a) Field dependence of magnetization curves for a CrI_3 crystal measured at ambient and 1 GPa pressure with the field parallel and perpendicular to the c axis at a representative temperature of 5 K. (b) Temperature dependence of zero-field-cooled (ZFC) magnetization for CrI_3 at different applied pressures with the magnetic field parallel to the c axis. The inset shows the pressure dependence of the two-step transition temperature T^* , determined from the position of the weak anomaly in $M(T)$ curves as shown by the arrow in the main panel.

shown for comparison. The magnetization under pressure also increases rapidly with increasing field and starts to saturate above 3 kOe, as in the case of ambient pressure with $H\parallel c$. Above 1 T, the value of M is close to that for ambient pressure with $H\parallel c$, and at around 2 T both curves merge with each other. This suggests that the Cr^{3+} remains in the high-spin state under pressure. However, a closer view reveals a significant difference between the $M(H)$ curves at ambient pressure and 1 GPa. In the low-field regime, the slope of the $M(H)$ curve, i.e., the magnetic susceptibility (dM/dH), is smaller for 1 GPa than that for the ambient pressure. The smaller value of dM/dH implies that the value of field required to achieve a state of saturation in magnetization increases with applied

pressure for $H\parallel c$. This may be due to the decrease in magnetic anisotropy under pressure in CrI_3 . The field dependence of M under pressure with $H\perp c$ could shed some light on this issue. This is, however, not possible with our current setup. If the anisotropy reduces under pressure, the $M(H)$ curves with $H\parallel c$ and $H\perp c$ should approach each other with increasing pressure. At low applied field, the $M(T)$ curve exhibits a weak anomaly slightly below T_C , which is more prominent in the zero-field-cooled cycle with $H\perp c$. This anomaly in M has been characterized as a two-step magnetic ordering but its origin is yet to be deciphered [39]. We have also recorded the temperature dependence of M in a zero-field-cooled cycle at different pressures with $H\parallel c$ to track this two-step transition at $T = T^*$, as shown in Fig. 3(b). The pressure dependence of T^* is shown in the inset of Fig. 3(b). Similar to T_C , T^* also increases sharply in the low-pressure region and then increases very slowly.

Mainly, two competing interactions are responsible for determining the magnetic ground state of CrI_3 . The direct exchange between Cr-Cr is AFM in nature. This direct exchange arises from the electron hopping between nearest-neighbor Cr sites and is maximum when the Cr-Cr bond angle, formed by the $3d$ orbitals, is 180° . On the other hand, depending on the symmetry relations and electron occupancy of the overlapping atomic orbitals, the superexchange can be FM as well as AFM in nature and is mediated through a nonmagnetic ligand ion. Unlike the direct exchange, the superexchange interaction originates due to the virtual hopping of electrons between the two nearest-neighbor Cr ions via an iodine ion. This virtual process reduces the total energy of the system. According to the Goodenough-Kanamori-Anderson rules, when the magnetic-ion-ligand-magnetic-ion angle is 90° , the superexchange interaction is FM. It is AFM when the angle is 180° [40–42].

In a weakly coupled layered van der Waals system, the effect of pressure on ferromagnetism is very sensitive to the bond angles and the interlayer coupling. If the magnetic-ion-ligand-magnetic-ion bond angle is 90° , T_C is expected to decrease with an application of pressure because pressure will either increase or decrease the bond angle from 90° . As a result, the superexchange interaction, which favors the FM ordering, weakens. On the other hand, T_C may increase with pressure, if the bond angle is lower or higher than 90° at ambient pressure. In such a situation, the bond angle may approach toward 90° with the application of pressure. It has already been mentioned that the effect of pressure on ferromagnetism, investigated on a bulk single crystal of $\text{Cr}_2\text{Ge}_2\text{Te}_6$ through magnetization measurements, is very different from what is observed in CrI_3 [33]. In $\text{Cr}_2\text{Ge}_2\text{Te}_6$, T_C decreases monotonically with pressure up to 1 GPa. First-principles calculations show that the Cr-Cr bond length decreases while the Cr-Te-Cr bond angle gradually diverges from 90° with increasing pressure. Both these effects favor a direct exchange and weaken the superexchange interaction, i.e., the AFM interaction in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ enhances under pressure. A theoretical calculation also shows that the c/a ratio decreases with pressure. The reduction of c is relatively more significant than that of a due to the weak interlayer interaction. As the interlayer coupling in CrI_3 is much weaker as compared to that in $\text{Cr}_2\text{Ge}_2\text{Te}_6$, the effect of pressure in increasing the coupling

between two adjacent layers is much more significant in the former. In bulk CrI_3 , the interlayer coupling (J) is FM in nature. Thus, one expects that T_C will increase with an increase in pressure due to the enhancement of interlayer FM coupling. Our $M(H)$ data under pressure also offer evidence of enhancements in interlayer coupling under pressure. In VI_3 , another ferromagnet with weak van der Waals coupling, T_C increases and the transition becomes sharp above a threshold value of P , which has been attributed to a crossover from two- to three-dimensionality due to the increase in interlayer coupling [35]. In order to understand why CrI_3 exhibits a highest T_C among the CrX_3 family and the effect of pressure on the FM transition in CrI_3 is different from that in CrBr_3 , we first briefly discuss the evolution of magnetism in CrX_3 with the size of the halogen ion X . As the Cr-Cr distance increases with increasing halogen size from Cl to Br to I, the direct exchange weakens. Also, by moving from Cl to Br to I, the covalent nature of the Cr- X bond enhances, which further strengthens the superexchange interaction as well as the spin-orbit coupling and hence enhances the FM ordering temperature [23]. Recent experiments suggest that the covalent nature of the Cr-I bond plays an important role in CrI_3 to engineer the highest T_C [43].

In order to take into account the anisotropic nature of the FM superexchange interaction via Cr-I-Cr, Lado and Fernandez-Rossier used an anisotropic XXZ -type Hamiltonian with an additional term in the Heisenberg model, the anisotropic symmetric exchange (λ), and estimated T_C from the spin-wave theory [23]. They observe that T_C increases with an increase in λ . In fact, the nature of the variation of T_C with λ is very similar to what we observe here from the pressure dependence. Another factor that may play an important role to increase the T_C in the CrI_3 system is the Cr-I-Cr bond angle. In CrI_3 , this bond angle is about 95° [22,23]. With an increase in pressure, the Cr-I-Cr bond angle may approach toward 90° . We believe that the different role of pressure on the FM transition of CrI_3 and CrBr_3 is partly due to their structural anisotropy and partly due to the strong covalent nature of the Cr-I-Cr bonding. A reduction in anisotropy with pressure also signifies a crossover from an Ising to a Heisenberg system. This leads to suppression in both T_C and susceptibility. On the other hand, as discussed earlier, the change in the bond angle and an increase in interlayer coupling J could give rise to enhancement in T_C under pressure. It appears, therefore, that the close competition between these two effects determines the nature of the pressure dependence of T_C and magnetization in different 2D magnetic systems. For example, while in CrI_3 we have observed a drop in low-field magnetization and nonlinear rise in T_C , the opposite behavior could be observed in CrBr_3 and other similar compounds. A high-pressure structural analysis may reveal important information for understanding the role of pressure on the FM interaction in the CrX_3 series.

In conclusion, we have studied the magnetic properties of the quasi-two-dimensional FM semiconductor CrI_3 under hydrostatic pressure. Unlike the two-dimensional FM semiconductors CrBr_3 and $\text{Cr}_2\text{Ge}_2\text{Te}_6$, in the present system, T_C is found to increase monotonically from 60.4 to 64.9 K as pressure increases from 0 to 1.0 GPa. With an application of pressure, the PM-FM transition becomes sharper whereas the

low-field magnetization decreases for a magnetic field parallel to the easy axis of magnetization. $M(H)$ data suggest that pressure makes the Cr^{3+} spins harder to orient along the c axis, and as a result, low-field magnetization decreases with an increase in pressure.

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