The effect of pressure on ferromagnetic properties of the van-der-Waals materials VI_3 and CrI_3

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Two-dimensional van-der-Waals (vdW) magnetic materials have in recent years become the subject of an intense research [1]. In these materials, hydrostatic pressure represents a powerful tuning parameter. The dominant effect of hydrostatic pressure on weakly bonded planes is consists of pressing them together which may gradually convert the system from two- to three-dimensional.

Despite belonging to a well-studied family of transitionmetal trihalides, the VI_3 and CrI_3 iodides have received a significant attention just recently [2, 3, 4, 5, 6]. VI_3 crystallizes in the trigonal *P31c* structure which reorders into a monoclinic *C*2/*c* structure below $T_S = 80$ K [2]. The material is a hard ferromagnet below $T_C = 50$ K with high anisotropy. Optical and electrical transport measurements reveal insulating properties and the previous theoretical predictions suggest VI_3 to be a correlated Mott insulator. The Curie temperature T_C has been reported intact by hydrostatic pressure up to ~ 0.7 GPa. The observed rapid increase of T_C at higher pressures up to 1 GPa has been attributed to the commencing departure of dimensionality away from two [2].

CrI_3, on the other hand, is a semiconductor which exhibits at T_C = 61 K a transition to an anisotropic 3D-Ising ferromagnetic state with the easy magnetization axis perpendicular to the layers [7]. The compound exhibits a large van der Waals gap which leads to a 3D magnetic characteristics. T_C increasing upon increasing pressure up to 1 GPa has been reported [8].

We present results comprehensive measurements of the magnetic properties of VI_3 and CrI_3 single crystals in hydrostatic pressures far exceeding the values reported sofar.

The single crystals were prepared by chemical vapor transport method as described elsewhere [3]. The reference ambient-pressure magnetization data with respect temperature and magnetic field was measured using PPMS systems (*Quantum Design*), and Closed Cycle Cryocooler (*Janis Research*), respectively. A double-layered CuBe/NiCrAl piston-cylinder pressure cell was used to generate pressures up to ∼3 GPa, with a Daphne 7373 pressure medium and a manganin manometer. Further extension of pressure-effect measurements up to 10 GPa using a DAC cell is in progress.

The temperature dependence of the real part of ac susceptibility χ_{Re} in VI_3 reveals clearly the ferromagnetic transition at ~ 50 K. Except of that, three additional, less pronounced peaks above T_C are observed in the temperature range of ~ 52 K-60 K. The anomalies seem to be almost unaffected by increasing pressure up to ~ 0.8 GPa. Above this pressure value, we observed the peaks merging into one and simultaneously T_C increases abruptly by 20% in 1.2 GPa. Similar pressure evolution of T_C was seen in Ref. [2]. For higher pressures up to 3.5 GPa, T_C increases linearly. The measured temperature dependence of magnetization reveals the ferromagnetic transition at ~ 50 K as well [2]; with increasing pressures, the transition becomes sharper and the absolute value of magnetization increases above ~ 0.6 GPa.

On the other hand, we have observed a significantly different pressure effect in CrI_3 compound, contradicting the results reported in [8]. Besides the dominant peak in the temperature dependence of the real part of AC susceptibility χ_{Re} corresponding to T_C, we observe another smaller peak at T^* ~ 50 K which shows identical pressure dependence. We observed only a very modest increase of T_C in low pressures up to 0.6 GPa which is not as significant as shown in Fig. 4 of Ref. [8]. The value of T_C does not change considerably in preasures up to ~ 1.5 GPa. At higher pressures, surprisingly, T_C starts to decrease. This linear decreasing tendency is observed up to the highest applied pressure of 3 GPa. The imaginary part of AC susceptibility vs. temperature χ_{Im}(T) shows peak only at T^* in all applied pressures. No frequency dependence was detected.

This different character of the pressure dependence of the ac susceptibility in the VI_3 and CrI_3 compounds is is tentatively attributed to different evolution of dimensionality of ferromagnetic order, respectively.

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