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FERROMAGNETIC SEMICONDUCTOR

# Toward intrinsic room-temperature ferromagnetism in two-dimensional semiconductors

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Two-dimensional (2D) ferromagnetic semiconductors have been recognized as the most promising candidates for next-generation low-cost, high-performance and nano-scale spintronic applications such as spin field-effect transistors and quantum computation/communication. However, as one of the 125 important scientific issues raised by Science journal in 2005 that "*is it possible to create magnetic semiconductors that work at room temperature*?", how to achieve a feasible ferromagnetic semiconductor with high Curie temperature is still a long-standing challenge despite of tremendous efforts have been devoted in this field since 1960s. The recent discovery of 2D ferromagnetic semiconductors $Cr_2Ge_2Te_6$ and $CrI_3$ has evoked new research interests in 2D intrinsic ferromagnetic semiconductors. But the low Curie temperature (< 45 K) of these materials is still badly hindering their industrial applications.

Recently, a group led by Professor Erjun Kan and Professor Hongjun Xiang gives a clue to solve this problem. They proposed that, by using two isovalent transition metal ions to construct an alloy compound, an intrinsic ferromagnetic semiconductor with Curie temperature up to room-temperature may be achieved. This chemical approach is based on a solid physical mechanism that, the superexchange interactions between adjacent magnetic ions could be enhanced by an on-site energy level staggering of *d* orbtials, as demonstrated by a simple double-orbital model. Because the exchange fields of different transition metal ions are usually different, the isovalent alloying may result in a large energy level staggering of *d* orbitals, leading to a significant enhancement of ferromagnetic couplings. They further used first-principles calculation methods to predict several double-metal ferromagnetic semiconductors, whose Curie temperatures are improved by 3–5 times in comparison with the single-metal basis materials. This work has revealed a new physical mechanism of enhancing ferromagnetic coupling in semiconductors without introducing any impurities or carriers and demonstrated the possibility of room-temperature ferromagnetic order in 2D semiconductors.

Fanlong Ning (Zhejiang University, Hanzhou, China)
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# Magnetism in $d^0$ oxides

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The emerging field of spintronics aims at the utilization of both the spin and charge of electrons, and the combination semiconducting and magnetic properties within a single material. Magnetic oxide semiconductors are significant spintronics materials. Besides of doping the semiconducting materials with a certain concentration of transition metals to integrate ferromagnetism into semiconductors, it is found that oxides of nonmagnetic cations exhibit elusive signs of weak temperature-independent ferromagnetism, namely "$d^0$ ferromagnetism". Up to now, a major research effort has been devoted to obtain ferromagnetism and explore the related mechanisms. Anyway, as the initial enthusiasm for magnetic oxides ebbed, with no encouraging sign of useful spintronics functionality, scientists tended to dismiss the magnetism as some sort of dirt effect or measurement artefact. Interest in unidentified magnetic objects waned. The publications on this subject declined rapidly.

Recently, in a comment, Professor J. M. D. Coey suggests that magnetism in $d^0$ oxides is associated with surface defects, but it defies conventional explanation. He outlines two hypotheses to explain $d^0$ magnetism, which are a spin-split defect impurity band and giant orbital paramagnetism related to zero-point vacuum fluctuations, respectively. He gives a few examples of $d^0$ oxides and points out the way forward including experiments designed to test which hypothesis, if either, is correct, and standard first-principles approaches to calculate electronic structure of surface defects, and band formation in the presence of natural adsorbates. At the same time, the practical implications are also mentioned.

Xiaohong Xu (Shanxi Normal University, Linfen, China)
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# Epitaxial lift-off of ferromagnetic (Ga,Mn)As nanoflakes for van der Waals heterostructures

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The recent discovery of two-dimensional (2D) van der Waals (vdWs) ferromagnetic crystals provides an ideal platform for fundamental understanding of 2D magnetism, as well as the applications of low-power spintronic devices. The advances of vdWs heterostructures can couple the quasiparticle interaction between the 2D ferromagnetic material and others with engineered strain, chemistry, optical and electrical properties, providing an additional route to realize conceptual quantum phenomena and novel device functionalities, such as unprecedented control of the spin and valley pseudospin, extremely large tunneling magnetoresistance, etc. However, due to their instability, the handing of 2D ferromagnetic materials can only be carried out under the help of encapsulation with other 2D materials in a glove box, which is the biggest barrier towards the practical applications.

Now, Kai Yuan and colleagues from Peking University, and Chinese Academy of Sciences, introduce an approach about peeling-off and transfer of 2D ferromagnetic (Ga,Mn)As layers

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with thickness of ~10–20 nm grown by the molecular-beam epitaxy under ambient conditions. The lifted-off freestanding (Ga,Mn)As layer maintained its ferromagnetism. Using mechanically layer-by-layer vdWs heterostructure assembly technique, they successfully fabricated vdWs heterostructure devices based on the lifted-off (Ga,Mn)As, including hBN/(Ga,Mn)As top-gate Hall device and p-(Ga,Mn)As/n-$MoS_2$ heterojunction diode. The electrical transport measurements demonstrated the ferromagnetic nature and gate tunable magnetoresistance of the lifted-off (Ga,Mn)As layer. The ambient stable lifted-off ultrathin (Ga,Mn)As layer can be used as an alternative 2D ferromagnetic materials.

Gang Xiang (Sichuan University, Chengdu, China)
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TWO-DIMENSIONAL MAGNETIC SEMICONDUCTORS

# Controlling magnetism in 2D $CrI_3$ by electrostatic doping

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Spin ordering in a semiconductor has attracted much attention in the community of condensed matter physics. By combining ferromagnetism and the semiconducting nature, systems such as Mn-doped PbSnTe or InAs were found to exhibit tunable magnetic properties responsive to an externally applied electric field. It thus holds great promises for developing novel spintronics with the tuning-knobs such as gate voltage. Conventionally, those systems are often studied in bulk forms and commonly referred to diluted magnetic semiconductor (DMS). In recent years, two-dimensional (2D) magnetism has become popular as spin interactions can prevail down to the monolayer limit in a number of gapped van der Waals materials, including the family of ferromagnetic $CrX_3$ (X = I, Br, Cl) and anti-ferromagnetic $MPS_3$ (M = Ni, Fe, Mn). It is of great importance to reconsider now the hunting for DMS by alternatively searching for an intrinsic magnetic semiconductor in the 2D magnetic systems.

Recently, researchers from Cornell University demonstrated that the 2D magnetic semiconductor $CrI_3$ exhibits strong tunability in its magnetic properties when subjected to an externally applied electrical field. It is known that bulk $CrI_3$ has a band gap of ~ 1.2 eV, and its bulk ferromagnetic Curie temperature is reported to be 64 K. When thinned down to the 2D limit, it turns into an anti-ferromagnetic coupling between layers, while spins are ferromagnetically coupled in an Ising manner in each layer. As a result, the net spin alignments become layer-dependent, i.e., ferromagnetic (FM) and anti-ferromagnetic (AFM) for odd and even number of layers, respectively. Strikingly, bilayered $CrI_3$ manifests a transform of magnetic hysteresis loop from AFM at hole doping to FM at electron doping. In other words, when zero external magnetic fields are applied, the magnetic ordering in bilayered $CrI_3$ can be shifted from AFM to FM, purely by electrostatic fields. The 2D $CrI_3$ has thus proven to be a platform for electrical control of spin, which opens up possibilities for future investigations of 2D intrinsic magnetic semiconductors.

In general, 2D magnetic semiconductors, being either ferromagnetic or antiferromagnetic, are building blocks for novel gate-tunable spintronic devices, such as magnetic pn junctions, interfacial topological magnetic ordering, flexible magnetic sensors/memories, and etc. Yet it is noticed that, so far, room temperature spin ordered semiconductor at the 2D limit are still rare, requiring ceaseless efforts from both experimental and theoretical sides.

Zheng Han (Institute of Metal Research, CAS, Shenyang, China)
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