Antiferromagnetic ordering in atomically thin 2-dimensional materials studied by Raman spectroscopy

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Magnetism in low dimensional systems is attracting much interest not only for the fundamental scientific interest but also as a promising candidate for numerous applications in sensors and data storage. However, most experimental studies on magnetism in 2-dimension so far have been limited to the magnetism arising from defects, vacancies, edges or chemical dopants which are all extrinsic effects. Recent discovery of ferromagnetism in atomically thin materials [1,2] ignited much interest in magnetism in 2 dimension in general. Antiferromagnetic ordering, on the other hand, is much more difficult to detect as the net magnetism is zero. Neutron scattering, which is a powerful tool to detect antiferromagnetic order in bulk materials, cannot be used for atomically thin samples. Raman spectroscopy has proven a powerful tool to detect ferromagnetic ordering by monitoring the zone-folding due to the antiferromagnetic order [3,4] or the signal from two magnon scattering. We report on the observation of *intrinsic* antiferromagnetic ordering in the two-dimensional limit. We demonstrate that $FePS_3$ exhibits an Ising-type antiferromagnetic ordering down to the monolayer limit, in good agreement with the Onsager solution for two-dimensional order-disorder transition. The transition temperature remains almost independent of the thickness from bulk to the monolayer limit with T_N ~118 K, indicating that the weak interlayer interaction has little effect on the antiferromagnetic ordering. [4] For an XXZ-type antiferromagnet $NiPS_3$, a signal due to two-magnon scattering and the low-energy scattering signal due to spin fluctuations are monitored to find antiferromagnetic ordering down to the monolayer limit. [5]

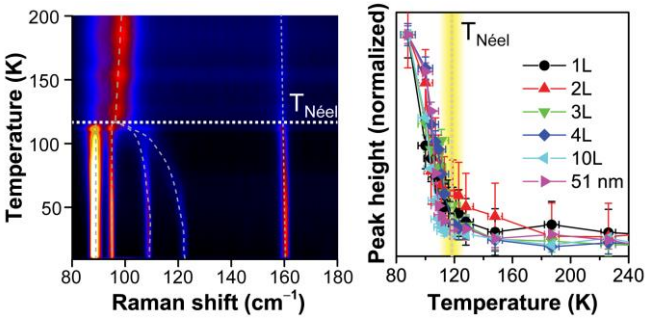

Figure 1. Evolution of Raman spectrum of $FePS_3$ (left) and comparison of Neel temperature for different thickness (right)

[1] C. Gong, *et al*., Nature **546**, 265 (2017).
[2] B. Huang, *et al.*, Nature **546**, 270 (2017).
[3] X. Wang, *et al*., 2D Materials **3**, 031009 (2016).
[4] J.-U. Lee, H. Cheong, *et al*., Nano Letters **16**, 7433 (2016).
[5] K. Kim, H. Cheong, *et al*., *in preparation*.

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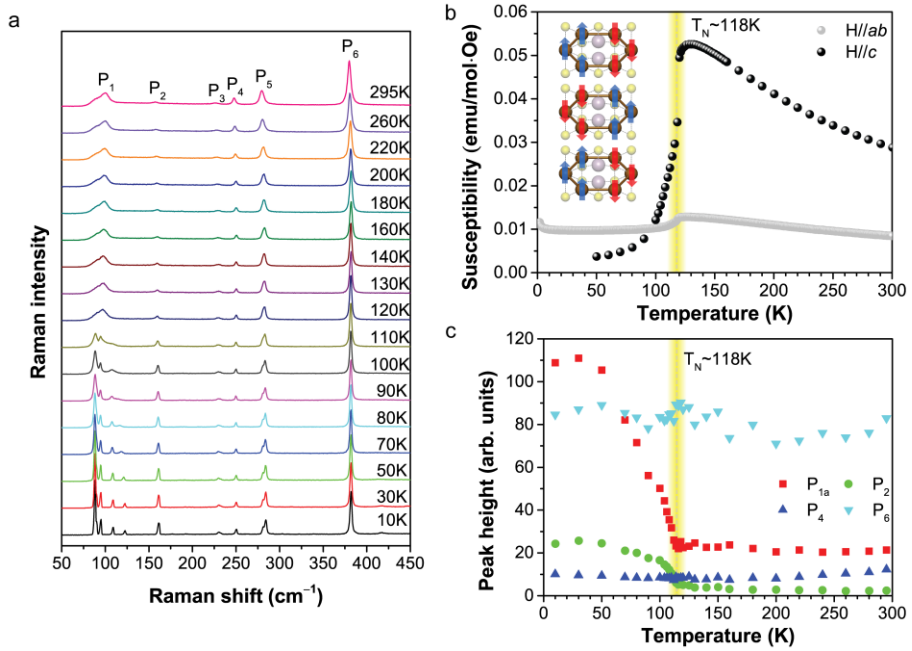

Figure 1. (a) Temperature dependence of Raman spectrum for bulk $FePS_3$. (b) Temperature dependence of magnetic susceptibility along *a* or *b* (black spheres) and *c* (grey spheres) axes. (c) Temperature dependence of intensities of several Raman peaks.

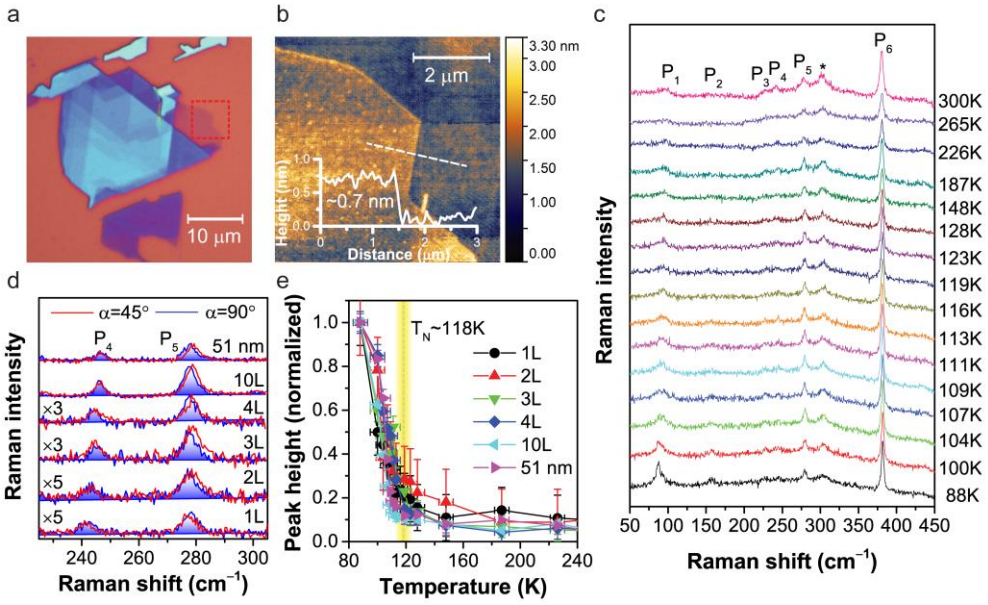

Figure 2. (a) Optical contrast and (b) atomic force microscope images of 1L $FePS_3$ on SiO_2/Si substrate. (c) Temperature dependence of Raman spectrum of 1L $FePS_3$ with polarization direction of $\alpha=\beta=45^o$. (d) Thickness dependence of P_3 and P_4 with polarization direction of $\alpha=45^o$ (red curves) and $\alpha=90^o$ (blue curves). (e) Temperature dependence of P_{1a} peak height for different thicknesses.