Quasi-two-dimensional magnon identification in antiferromagnetic FePS3 via magneto-Raman spectroscopy

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[Ques] Recently it was discovered that van der Waals bonded magnetic materials retain long range magnetic ordering down to a single layer, opening many avenues in fundamental physics and potential applications of these fascinating materials. One such material is FePS3, a large spin (S = 2) Mott insulator where the Fe atoms form a honeycomb lattice. In the bulk, FePS3 has been shown to be a quasi-two-dimensional-Ising antiferromagnet, with additional features in the Raman spectra emerging below the Néel temperature (TN) of approximately 120 K.

[关键问题] 作者试图回答：$FePS_3$ 在反铁磁有序态中新增的 Raman 模式是否来源于磁子而非声子？

[Sum] Using magneto-Raman spectroscopy as an optical probe of magnetic structure, we show that one of these Raman-active modes in the magnetically ordered state is actually a magnon with a frequency of ≈3.7 THz (122 cm−1). Contrary to previous work, which interpreted this feature as a phonon, our Raman data shows the expected frequency shifting and splitting of the antiferromagnetic magnon as a function of temperature and magnetic field, respectively, where we determine the g factor to be ≈2. In addition, the symmetry behavior of the magnon is studied by polarization-dependent Raman spectroscopy and explained using the magnetic point group of FePS3.

[亮点 1] 通过磁拉曼光谱首次明确证明 $FePS_3$ 在 Néel 温度以下出现的特征峰并非声子，而是频率约 3.7 THz 的反铁磁磁子，为二维范德华反铁磁体的磁激发研究提供了关键光学证据。

[亮点 2] 利用温度依赖与磁场依赖的 Raman 位移/分裂行为提取磁子 g 因子（≈2），并结合偏振光谱揭示其受磁点群约束的对称性，实现对磁子性质的完整光学表征与机制解释。

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[思考] （1）$FePS_3$ 的磁子在超薄层甚至单层中是否仍保持 Raman 活性？其维度降低后能否展现更强的量子涨落或拓扑磁激发特征？

[思考] （2）若向 $FePS_3$ 中施加应变、电场或异质结构耦合，其磁子能谱是否会出现可调谐带隙或模式混合，为自旋波器件设计提供新可能？

[拓展阅读 1] Raman-active magnon 与 Raman-active phonon 的区别是什么？在 Raman 测试中的表现有何不同？

[拓展阅读 1] 在固体中， phonon（声子）代表晶格原子的集体振动，而 magnon（磁子）则代表自旋体系中的集体激发（即自旋翻转或自旋波）。两者都可能在 Raman 光谱中产生可观测峰，但其物理起源和光谱特征明显不同。

首先，声子由晶格振动决定，因此其能量主要受晶体结构、化学键强度和质量分布影响，与磁场的直接耦合极弱。因此，在 Raman 实验中，声子峰在磁场下通常不会发生明显移位或分裂，只有温度引起的热膨胀或 anharmonic 效应会导致轻微变化。相比之下，磁子反映的是自旋之间的交换作用和磁序结构，因此对 温度、磁场和磁对称性高度敏感。例如：在温度降低进入磁有序相后，磁子模式才出现或增强；外加磁场会改变自旋能级，导致磁子峰发生频率移动（Zeeman shift）甚至分裂； 偏振 Raman 测试中，磁子是否出现以及强度如何，取决于磁点群和 Raman 张量的选择定则。因此，在 Raman 光谱中，当某峰表现出明显的磁场依赖性、温度依赖性以及偏振对称性特征时，它更可能是磁子而非声子。$FePS_3$ 的这篇工作正是利用这些差异确定了 3.7 THz 模式的磁性本质，为二维反铁磁系统的光学自旋激发研究提供了范例。

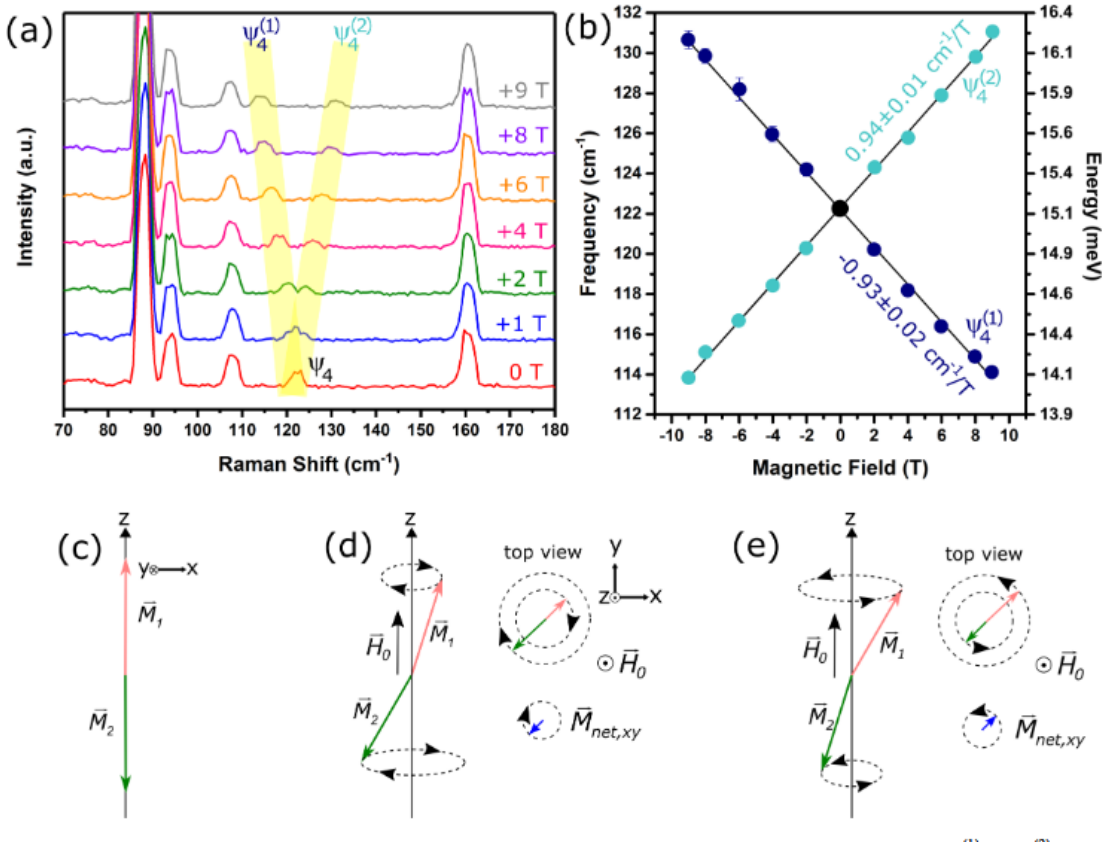
[introduction] Since the isolation of monolayer graphene in 2004 [1], there has been a surge of research into van der Waals layered materials, where the strong intralayer and weak interlayer coupling allows for isolation of layers that are only a few atoms thick. These materials exhibit a wide range of electronic properties, including semiconducting, metallic, insulating, superconducting, and charge density waves, allowing for device architectures composed of solely two-dimensional (2D) materials [2–4]. While significant research has been dedicated thus far to investigating the optical, mechanical, and electrical properties of 2D materials [3,5–9], exploring magnetism is still in its infancy, even though 2D magnetic materials provide a solid-state platform to experimentally access fundamental, low-

dimensional physics [10,11]. Additionally, any 2D magnetic material would likely still possess the captivating properties of 2D materials, including extremely large mechanical flexibility [12,13], efficient tuning of transport properties with an electric field [14–17], relative ease of chemical modification [18,19], as well as the ability to create van der Waals stacked heterostructures [20]. These myriad of tuning parameters could unlock opportunities for custom-engineered magnetoelectric and magneto-optical de vices, where 2D magnets coupled with other technologically relevant materials could realize unprecedented capabilities in fields such as spintronics [11,21,22].

In early 2017, intrinsic ferromagnetism was observed down to the few-layer and monolayer limit in two different, layered materials with magnetic anisotropy: Cr2Ge2Te6 and CrI3 [23,24]. For CrI3, it was also shown that the interlayer magnetic ordering (i.e., ferromagnetic or antiferromagnetic stacking) was dependent on the number of layers [24], and could be controlled by an external electric field [14,25]. Transition metal phosphorus trisulfides XPS_3 (X=Fe, Mn, Ni, etc.) are another class of van der Waals antiferromagnets that are being studied in the 2D limit. Interestingly, although FePS3 (TN ≈ 120 K) [26], MnPS3 (TN ≈ 78 K) [26], and NiPS3 (TN ≈ 155 K) [26] are isostructural, they have different spin structures below the N é el temperature. The choice of transition metal results in varied magnetic phenomena, since the spins align antiferromagnetically within the layers in different fashions, including N éel, zigzag, or stripe ordering [27,28]. FePS3, which is a Mott insulator [29,30], is especially intriguing as a 2D Ising antiferromagnet on a honeycomb lattice [26,31]. In addition, long-distance magnon transport (several micrometers) has been experimentally observed in MnPS3, demonstrating that these materials are viable candidates for possible magnonic devices [22].

Raman spectroscopy, being nondestructive and highly sensitive to minute lattice perturbations, is a powerful technique to study various properties of quantum materials, including the effects of layer number [32,33], strain [34], defects/doping [35], electron-phonon coupling [36,37], phase transitions [38], spin-phonon coupling [39], and magnetic excitations [40,41]. In addition, unlike other measurements that require bulk, large-area crystals, such as neutron diffraction, x-ray diffraction, or magnetic susceptibility, Raman spectroscopy can probe atomically thin flakes with diffraction-limited spatial resolution (e.g. micrometer spot size). The Raman spectra of bulk X PS3 materials have been studied previously [27,42,43], and only recently extended to samples in the monolayer limit for

NiPS3 [44] and FePS3 [45,46]. In particular, FePS3 is an interesting candidate to study using Raman spectroscopy because, due to the antiferromagnetic alignment of the spins and the resulting increase in the unit cell, Brillouin zone folding leads to new modes appearing below TN [45,46].



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