Probing magnon-phonon coupling in van der Waals antiferromagnets by Magneto-Raman scattering

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The excitations of the lattice and the magnetic degrees of freedom as separate subsystems have been extensively studied experimentally and theoretically over recent decades. They have been described both qualitatively and quantitatively in many crystalline materials. In the next step, the focus has shifted towards understanding the interplay between lattice vibrations and spin excitations. Phonons and magnons can interact through magnetoelastic coupling, giving rise to hybridized collective spin-lattice modes known as magnon-polarons. This hybridization offers potential mechanisms for exciting, manipulating, and detecting magnons, which are key requirement for magnonics. Previous studies demonstrate the coupling between acoustic phonons and magnons in ferromagnetic materials, whose intrinsic magnon energies are typically in the GHz range [1]. In recent years, antiferromagnetic (AFM) based magnonics is rapidly emerging. The AFM magnons exhibiting energies in the THz range due to exchange and finite anisotropy energy. This high magnon energy coincides with the energy of optical phonons, indicating a possibility of magnon-phonon interaction. It has been evidenced in recent work where the effects of magnon-phonon interaction were monitored by tuning the magnon (phonon) energy through external magnetic field or pressure [2,3].

In this study, we employ Raman scattering technique under various environments of temperature, magnetic field, and hydrostatic pressure to investigate the magnon and magnonphonon coupling in antiferromagnets from the MPX₃ (M=Ni, Mn, Fe, Co; X=S, Se) family. We observe the presence of magnon mode(s) near the optical phonon modes originating from the vibration of the metal atoms. In the case of FePS₃, magnon and phonon are uncoupled at low temperatures, but a change in temperature, external field, or pressure induces the formation of magnon-polarons [2,3]. Conversely, in FePSe₃, the magnon-polaron coupled mode is present even at zero field and pressure. Applying an external magnetic field helps to disentangle the coupled excitations and identify the bare magnon gap [4]. In other materials, we observe coupling between phonons and the two-magnon continuum, resulting in a Fanoshaped phonon mode. Through these measurements, we determine coupling constants, the type of coupling (selective or non-selective), bare magnon modes, and the effective g-factor. Additionally, we discuss the presence of an in-plane anisotropy in FePSe3 and interlayer exchange interactions in the antiferromagnets.

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