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Acoustic and optical phonons in quasi-two-dimensional MPX_3 antiferromagnetic semiconductors

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ABSTRACT

We report the results of the investigation of the acoustic and optical phonons in quasi-two-dimensional antiferromagnetic semiconductors of the transition metal phosphorus trisulfide family with Mn, Fe, Co, Ni, and Cd as metal atoms. The Brillouin–Mandelstam and Raman light scattering spectroscopies were conducted at room temperature to measure the acoustic and optical phonon frequencies close to the Brillouin zone center and the $\Gamma - A$ high symmetry direction. The absorption and index of refraction were measured in the visible and infrared ranges using the reflectometry technique. We found an intriguing large variation, over $\sim 28\%$, in the acoustic phonon group velocities in this group of materials with similar crystal structures. Our data indicate that the full-width-at-half-maximum of the acoustic phonon peaks is strongly affected by the optical properties and the electronic bandgap. The acoustic phonon lifetime extracted for some of the materials was correlated with their thermal properties. The results are important for understanding the layered van der Waals semiconductors and assessing their potential for optoelectronic and spintronic device applications.

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Recently, low-dimensional magnetic semiconductors have become a focus of theoretical and experimental investigation as a new group of materials with potential applications in spintronic devices.^{1–9} Following the discovery of graphene, a two-dimensional (2D) single-atomic sheet of carbon atoms, interest was directed to magnetic graphene-like structures that host different forms of spin order even at few and single-layer limits.^{2,3,10} Metal-transition phosphotrichalcogenides, referred to as MPX_3 in which M is a transition metal and X is a chalcogen, are a class of compounds that attracted much attention. Thin films of many of such materials with the few-atomic-plane and the single-atomic-plane thicknesses reveal antiferromagnetic (AFM) spin configurations, below their respective Néel temperatures, T_N .^{11–16} Theoretical predictions suggest that quasi-2D MPX_3 materials have spin order tunability via external stimuli such as strain fields and electric gating.^{11,17} The latter combined with their low cleavage energy present opportunities for the fabrication of magnetic heterostructures

with the potential for electronically controlled spintronic technologies.^{18,19} While the properties of AFM electrical conductors and insulators are well known, the AFM semiconductors are still poorly understood. Relatively, little is known about the interplay of the electron, magnon, and phonon properties of such materials.

Early interest in MPX_3 compounds was motivated by their prospects for applications as cathodes in lithium batteries with research focused on characterization of the material properties in their bulk form.²⁰ There are published reports on bulk magnetic and electrical properties of MPX_3 s.^{21–23} Although there were prior Raman spectroscopy studies of optical phonons in MPX_3 materials,¹⁸ the information on the acoustic phonon properties of MPX_3 materials is missing.²⁴ Acoustic phonons carry heat and limit the electron mobility near room temperature in semiconductor materials. The lattice thermal conductivity, K_P , of semiconductors is directly related to the group velocity of acoustic phonons, v , via $K_P \sim C v \Lambda$, where C and Λ

represent the volumetric specific heat and phonon mean free path, respectively. The characteristics of the long-wavelength acoustic phonons are defined by the elastic coefficients of the media.²⁵ The latter is particularly important, considering that the magnetic states of MPX_3 films can be engineered through strain fields.^{11,19,26} The practical value of the knowledge of acoustic phonons not only is limited to heat transport, scattering of electrons, and determining mechanical properties but also extends to the understanding of magnon–phonon scattering. Theoretical studies have shown that the interplay between low-energy magnons and long-wavelength acoustic phonons close to the Brillouin zone (BZ) center influences the magnon relaxation processes.²⁷

In this study, we focus on the important sub-group of MPX_3 materials, namely, MPS_3 . We report the results of the investigation of the acoustic and optical phonons in a set of MPS_3 compounds with Mn, Fe, Co, Ni, and Cd transition metal atoms. We aim to determine the phonon characteristics of the AFM semiconductors important for understanding their properties. The questions we ask are: How do the details of the crystal structure affect the acoustic and optical phonon spectra in this type of material? Can we observe any signatures of the interplay of the electronic, phononic, and spin properties of these materials using optical spectroscopy tools? To achieve this, we employ micro-Brillouin–Mandelstam light scattering (BMS) and micro-Raman techniques to detect acoustic and optical phonons, respectively. BMS is a nondestructive optical technique that is used to detect elemental excitations such as acoustic phonons and magnons with energies in the range from 2 to 900 GHz and wavevectors close to the BZ center.²⁸ The optical properties of MPS_3 films were measured via spectral reflectometry in the visible light range. The data on the optical properties are required for the interpretation of the BMS spectra and for extracting the group velocity of acoustic phonons. The high-quality bulk single crystals of MnPS_3 , FePS_3 , CoPS_3 , NiPS_3 , and CdPS_3 were synthesized using the chemical vapor transition (CVT) method (2D Semiconductors, USA). Raman and BMS measurements were performed on bulk crystals at room temperature (RT). The crystals were mechanically exfoliated on Si/SiO₂ substrates for optical reflectometry measurements. The thickness of the exfoliated samples ranged about a few hundred nanometers, while their lateral dimensions typically varied between 10 to 30 μm .

Figure 1 presents a schematic of the different magnetic order and crystal structures of the studied materials. The spin arrangement in these semiconductors is primarily influenced by the size of the “M” elements, which are arranged in hexagonal sublattices. The diverse ionic radii of different transition metal cations lead to various AFM spin configurations, such as Néel, zigzag Ising, or zigzag XY spin ordering.²⁹ Figure 1(a) illustrates different magnetic arrangements observed in different compounds within this family. The small ionic radii for the Co^{2+} and Ni^{2+} cations result in flat octahedral structures around the transition metal, where spins are constrained from freely rotating about the ab plane.²⁹ In contrast, the larger radii of Fe^{2+} and Mn^{2+} cause the spins to rotate about the ab plane and align along the c -axis of these compounds. As a result, MnPS_3 and FePS_3 exhibit out-of-plane spins with Néel and zigzag Ising AFM alignment, respectively, while CoPS_3 and NiPS_3 both have zigzag XY ordering with spins oriented within the ab plane.²⁹ Other compounds with the metal elements of Cd and Zn are nonmagnetic. The effect of ionic radius can be seen in the Néel temperature as well. Compounds with the smallest cation radius and lattice constants show the highest Néel

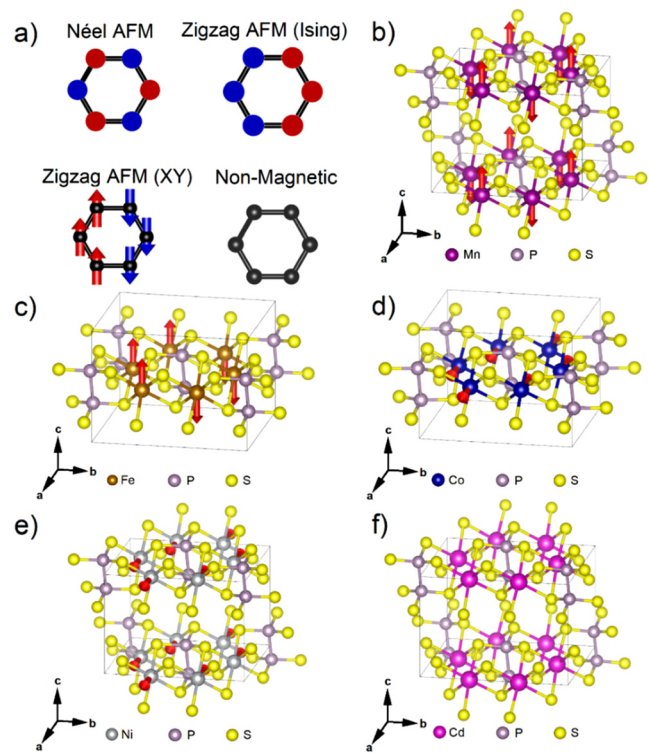


FIG. 1. (a) Schematic showing different magnetic configurations of MPS_3 semiconducting compounds. The blue and red spheres represent opposite spin ordering perpendicular to the ab plane. The arrows indicate the opposite spin ordering within the ab plane. The crystal structures of various MPS_3 s with metal atoms of (b) Mn, (c) Fe, (d) Co, (e) Ni, and (f) Cd. The arrows show the spin orientation configuration in different crystals.

temperatures.^{11,29} All studied MPS_3 s have a monoclinic structure, comprised of three unequal axis lengths with two being perpendicular and the third at an oblique angle to the perpendicular axes. They all have twofold, body-centered mirror symmetry corresponding to the $C/2m$ space group. The sulfur atoms bond with phosphorus to form dumbbell-like units of P_2S_6 . Figures 1(b)–1(f) present the crystal structure and spin alignment of different MPS_3 compounds. Table I summarizes the lattice parameters, Néel temperatures, and AFM type of each compound used in this study.

Raman spectroscopy was performed in the conventional back-scattering configuration using two laser excitation wavelengths of 488 nm (blue) and 633 nm (red) at RT. The Raman filter cutoff wavenumber for both blue and red lasers was $\sim 110\text{ cm}^{-1}$. In all experiments, the incident light was perpendicular to the (001) surface. The polarization of the scattered light was not analyzed. To avoid any local heating effects, the excitation laser power was kept below $\sim 500\text{ }\mu\text{W}$ in all measurements. Figures 2(a) and 2(b) show the Raman spectra of all samples under 488 and 633 nm laser excitations, respectively. Note that at RT, all studied MPS_3 s are in the paramagnetic phase, and therefore, they belong to the C_{2h} point symmetry group with the irreducible representation of $\Gamma = 8A_g + 6A_u + 7B_g + 9B_u$. Among these modes, A_g and B_g modes are Raman-active, which are assigned to their respective Raman peaks in Figs. 2(a) and 2(b).²² Note that the symmetry of

TABLE I. Structural, electronic, and magnetic properties of MPS₃ compounds.

	a (Å)	b (Å)	c (Å)	β (°)	Bandgap (eV) ^a	Spin order	T _N (K)	References
MnPS ₃	6.077	10.524	6.796	107.35	3.0 (D)	Néel AFM	78	11, 12, 30, and 31
FePS ₃	5.947	10.300	6.7222	107.16	1.5 (I)	Zigzag (Ising)	118	10–12
CoPS ₃	5.901	10.222	6.658	107.17	1.6 (I)	Zigzag (XY)	132	11, 12, and 32
NiPS ₃	5.812	10.070	6.632	106.98	1.4 (I)	Zigzag (XY)	155	11, 12, and 33
CdPS ₃	6.218	10.763	6.867	107.58	3.3 (I)	NM	NM	11 and 12

^a“D” and “I” stand for direct and indirect bandgap materials, respectively.

these compounds would be different in their monolayer form below their magnetic transition with all nonmagnetic and Néel antiferromagnetic types belonging to *D*_{3d} and the zigzag AFMs preserving their *C*_{2h} point symmetry groups.¹²

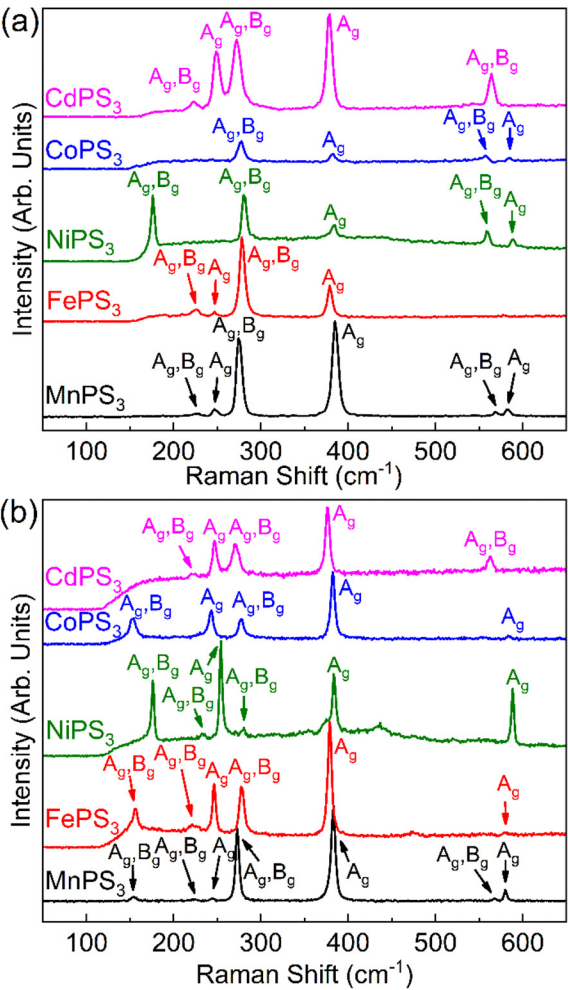


FIG. 2. Raman spectra of MPS₃ layered semiconductor compounds at room temperature using different laser excitations of (a) 488 nm and (b) 633 nm with the vibrational mode symmetries assigned.

It is important to mention that not all *A_g* and *B_g* modes were detectable in the spectra measured for all compounds, either due to their weak intensities or Raman filter cutoff limitations in the low-wavenumber range. For example, we were not able to detect the expected *A_g* mode at ~580 cm⁻¹ for CdPS₃. The frequency and the full-width-at-half-maximum (FWHM) of the Raman peaks along with their mode assignments are listed in Table II. The data were obtained using 633 nm (red) and 488 nm (blue) laser excitations. It is generally considered that despite the complex atomic structure of MPS₃ materials, their vibrational characteristics can be pictured into isolated high-wavenumber vibrations of P₂S₆⁴⁻ and lower-wavenumber vibrations of M²⁺ and P₂S₆⁴⁻ with spectral features above and below ~200 cm⁻¹, respectively.^{22,24} As a result, the spectral position of the peaks in the high wavenumber ranges remains largely invariant across structures with different M atoms.²² In contrast, the spectral position of the peaks at low wavenumber ranges is influenced by the transition metal. While the degenerate *A_g*, *B_g* modes for MnPS₃, FePS₃, and CoPS₃ are observed at ~155 cm⁻¹, it is located at 176 cm⁻¹ for NiPS₃. The reason for such a large spectral shift given the almost identical atomic masses of Mn, Fe, Ni, and Co is most likely related to a smaller cation radius of Ni²⁺ and requires theoretical calculations. The atomic mass of Cd is almost twice that of the other transition metals of interest in this study, and, thus, the same degenerate peaks are expected to be situated at ~110 cm⁻¹. Prior Raman studies of CdPS₃ confirm degenerate *A_g*, *B_g* modes at or close to the calculated wavenumber.³⁴ It is worth noting that certain reports attribute the Raman peaks of MPS₃ structures in accordance with the *D*_{3d} point group symmetry due to the weak inter-layer weak vdW bonds, with *E_g* and *A_{1g}* modes being Raman active.³⁴ This assumption has led to inconsistencies in Raman mode assignment across the literature. However, we contend that, considering the *C*_{2h} point symmetry of these structures, Raman-active modes should be designated as *A_g* and *B_g*.

The optical properties, i.e., the index of refraction, *n*, and extinction coefficient, *k*, of the samples were determined using spectral reflectometry (Filmetrics F40, KLA Instruments, USA). In these experiments, the samples were illuminated with a tungsten-halogen light source, and the reflectance was measured as a function of the wavelength in the range of 400–1000 nm. The measurements were performed with the incident light normal to the sample surface. The spot size on the sample was ~8 μm. Fittings were performed to determine the refractive index and optical extinction coefficient as a function of wavelength. The results for *n* and *k* are presented in Figs. 3(a) and 3(b). The examined materials show a general trend of decreasing refractive index with the wavelength. Semiconductors with smaller band gaps exhibit higher refractive indices, as expected.³⁵ It is known

TABLE II. Frequency, FWHM, and symmetries of Raman peaks MPS₃ compounds.

Symmetry			A _g , B _g	A _g , B _g	A _g	A _g , B _g	A _g	A _g , B _g	A _g
MnPS ₃	Frequency	Red	154.1	223.7	244.6	273	382.8	567.6	580.2
		Blue	...	226.4	247.5	275.2	385.2	569.3	583
	FWHM	Red	6.5	5.6	4.3	5.7	6.6	6.3	4.3
		Blue	...	7.5	6.7	8	9.1	7.5	8.1
FePS ₃	Frequency	Red	155.8	223.2	246.4	278	378.8	...	580.2
		Blue	...	225.2	246.7	278.5	379.0	...	...
	FWHM	Red	7.4	11.4	4.9	6.9	6.3	...	5.8
		Blue	...	9.7	3.2	8.0	7.3	...	...
NiPS ₃	Frequency	Red	176.1	233.2	254.2	279.8	383.7	...	588.4
		Blue	176.1	...	...	280.9	383.3	559.4	588.8
	FWHM	Red	3.8	4.7	4.0	4.0	6.3	...	3.8
		Blue	5.4	...	...	6.6	7.8	6.1	5.2
CoPS ₃	Frequency	Red	153.1	...	242.6	277.4	382.3	...	583.9
		Blue	...	...	...	277.2	382.1	556.0	584.5
	FWHM	Red	9.9	...	8.4	8.1	6.4	...	4.6
		Blue	...	...	...	9.4	7.3	10.2	5.1
CdPS ₃	Frequency	Red	...	221.9	247.1	271	376.3	562.5	...
		Blue	...	222.9	249.4	272.2	378.4	564	...
	FWHM	Red	...	8.6	6.2	9.4	6.5	6.8	...
		Blue	...	10.7	10.0	12.9	8.4	7.8	...

that MPS₃s are semiconductors with bandgaps spanning the mid-range of 1.3 eV to a wide-bandgap range of 3.5 eV (see Table I).³⁵ All structures in this study are indirect bandgap semiconductors except for MnPS₃, which is a direct bandgap semiconductor. The data on the optical properties at 532 nm excitation wavelength are required for the interpretation of BMS spectra, which will be discussed next.

We now turn to the main element of this work, which is the investigation of the acoustic phonon energies in MPS₃ compounds using the BMS technique. The BMS measurements were conducted in the backscattering configuration using a continuous wave diode-pumped laser (Spectra-Physics, Excelsior, USA) with an excitation wavelength of $\lambda = 532$ nm. The incident laser power was measured at ~ 10 mW at a constant incident angle of $\theta = 45^\circ$ on all samples. The incident and scattered light were both *p*-polarized. The details of the BMS setup and experimental procedure can be found in our prior reports for other material systems.^{36–38} The results are presented in Fig. 4. The spectra are vertically transferred for better visualization. Two sharp peaks on both sides of the spectra, corresponding to the Stokes and anti-Stokes processes, were observed in the frequency range of ± 27 to ± 35 GHz, for all samples. These peaks depicted by “LA” in the plot are attributed to longitudinal acoustic phonons propagating close to the *c* crystallographic axis in the real space or the $\Gamma - A$ direction in the reciprocal space, for all structures. Symmetrical spurious peaks were also observed in the Stokes and anti-Stokes sides of the spectra that are shaded by gray rectangles for clarification. These peaks likely originated from the optical cavity

formed by the reflection of the light from the front and back surfaces of the samples.

The spectral frequency, *f*, and the FWHM of the LA peaks were determined by fitting individual Lorentzian functions to the experimental data. The obtained frequencies for MPS₃s with Mn, Fe, Co, Ni, and Cd transition metal atoms are 33, 34, 37, 33, and 27 GHz, respectively. Note that, unlike optical phonons which typically have a flat dispersion in the vicinity of the Brillouin zone center, acoustic phonons have a linear dispersion as $\omega = qv$ close to the Γ point, in which $q = 4\pi n/\lambda$ is the phonon wavevector, $\omega = 2\pi f$ is the angular frequency, and *v* is the phonon group velocity. Using the dispersion relation, the group velocity can be calculated according to $v = f\lambda/2n$. We obtained a group velocity of 2960, 2713, 3203, 2310, and 3077 m/s for the Mn, Fe, Co, Ni, and Cd-based compounds, respectively. The results show that longitudinal acoustic phonons propagate with rather slow velocities in the range of ~ 2300 to ~ 3200 m/s for MPS₃ compounds, with NiPS₃ and CoPS₃ exhibiting the slowest and fastest propagation velocities, respectively. The obtained results are in acceptable agreement with limited studies investigating the acoustic phonon properties in layered materials.³⁹ One prior Brillouin spectroscopy study of the acoustic phonons in CdPS₃ reported a peak associated with LA phonons propagating close to the *c* crystallographic direction at 28 GHz, which is consistent with our results.³⁹ However, the reported *v* in that study was slightly lower than our value owing to the higher *n*, which was taken from the literature.

An intriguing observation in the spectra presented in Fig. 4 is a substantial disparity among the FWHM of the LA peaks for different

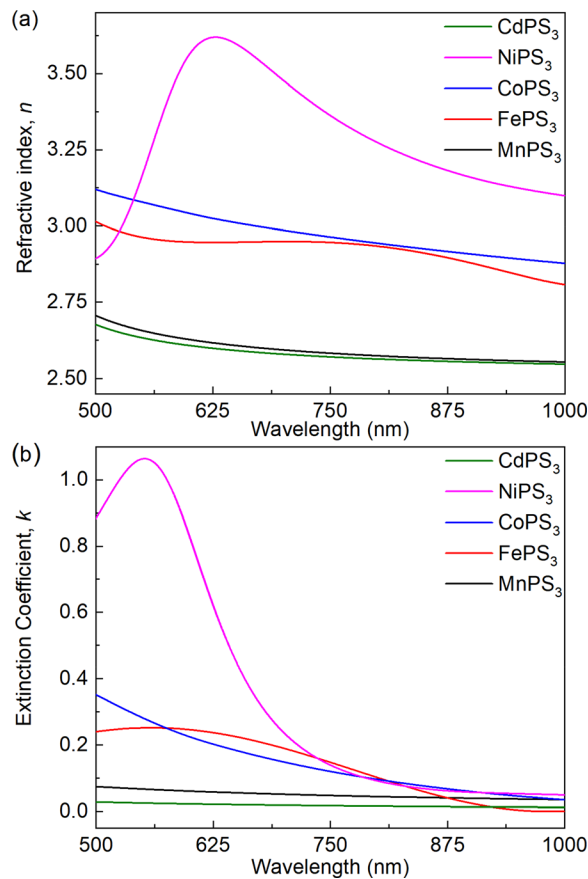


FIG. 3. (a) Index of refraction, n , and (b) extinction coefficient, k , of MPS₃ compounds at room temperature.

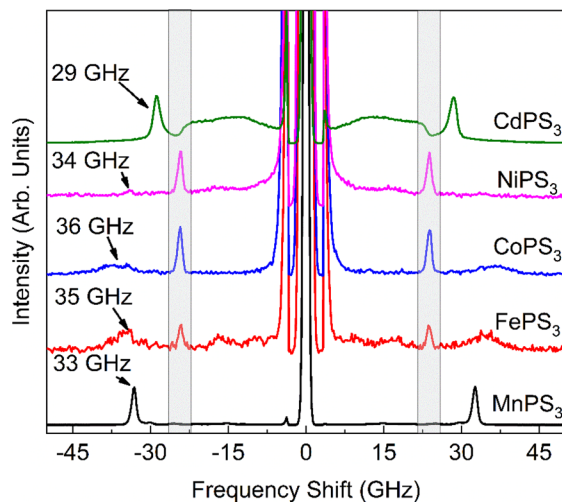


FIG. 4. BMS spectra of MPS₃ compounds performed at room temperature at an incidence angle of 45°. Peaks shown with arrows are associated with the longitudinal acoustic (LA) phonon modes.

compounds. The large FWHM of the BMS peaks in specific compounds is a direct manifestation of high optical absorption in those structures.⁴⁰ In fact, as seen in Fig. 4, the LA peaks in the spectra accumulated for Fe, Ni, and Co-based structures are so broad that they are barely detectable even after long accumulation times. For an opaque or a semi-transparent crystal with the complex refractive index $\eta = n + ik$, an optical broadening of $\delta\omega/\omega = 2k/n$ occurs due to the scattering of light from phonons with momentums within $\delta p \sim 2kk_0\hbar$ and mean phonon momentum of $p = q\hbar$ due to the uncertainty principle.⁴⁰ For Fe, Ni, and Co-based structures, taking the measured values of n and k , the optical broadening is calculated as ~ 5.7 , 22.5 , and 7.2 GHz, respectively. However, the optical broadening for semi-transparent materials of MnPS₃ and CdPS₃ at $\lambda = 532$ nm is ~ 1.7 and 0.6 GHz, respectively. The experimental FWHM of the LA peaks obtained from individual Lorentzian fittings on the BMS data yields ~ 1.0 , ~ 6.5 , ~ 18 , 1.6 , and 1.7 GHz for Mn, Fe, Co, Ni, and Cd-based structures, respectively. Comparing the experimental and calculated optical broadening values, one would infer that only in the case of CdPS₃, the optical broadening is significantly lower than the obtained experimental FWHM. In this specific case, the FWHM can be related to the phonon lifetime considering the energy-time uncertainty relation $\Delta E/\hbar = 1/\tau$ in which ΔE , $\hbar$, and τ are the BMS peak FWHM (rad/s), reduced Planck's constant, and the phonon lifetime, respectively. A phonon lifetime of ~ 144 ps is derived after subtracting the optical broadening of 0.6 GHz from the measured 1.7 GHz FWHM for the probed LA phonon in CdPS₃. The obtained phonon lifetime is rather short for such long-wavelength acoustic phonons with wavevectors close to the BZ center and frequency of 28 GHz. To provide a comparison, the reported lifetime of 50 – 100 GHz LA phonons in a silicon wafer is ~ 5 – 7 ns, while for 30 GHz LA phonons in free-standing silicon membranes is ~ 2 ns.^{41,42} The slow phonon group velocity and short phonon lifetime of these structures imply a poor cross-plane thermal conductivity of these compounds. A prior experimental study reports significantly low cross-plane thermal conductivity values of 0.85 and 1.1 W/mK for FePS₃ and MnPS₃, respectively, which aligns with our findings on acoustic phonon properties.²⁴

At cryogenic temperatures, long-wavelength acoustic phonons are the main heat carriers in semiconductor materials. Lattice thermal conductivity, $K_p \sim C v \Lambda$, is directly related to the phonon group velocity. An intuitive method to verify the accuracy of group velocities obtained through BMS measurements is to examine the thermal conductivity of these compounds at cryogenic temperatures. Such data are available for FePS₃, MnPS₃, and NiPS₃.⁴³ Our findings show that the group velocity of NiPS₃ is lower than that of FePS₃, which, in turn, is slightly less than that of MnPS₃. Interestingly, existing thermal transport data also support this trend, showing that NiPS₃ and MnPS₃ have the smallest and largest thermal conductivity values, respectively. Our results on the acoustic phonons, i.e., phonon group velocities and the phonon lifetime, correlated with the available thermal data, suggest that the AFM semiconductors would require special thermal management provisions for practical applications. The latter may include placing MPS₃ on thermally conductive substrates, e.g., diamond, in a similar way as it is done for ultra-wideband-gap Ga₂O₃ semiconductors characterized by low thermal conductivity.

In conclusion, we reported the results of the investigation of the acoustic and optical phonons in quasi-2D layered semiconductors of the MPS₃ family with Mn, Fe, Co, Ni, and Cd as metal atoms.

All examined materials, except one CdPS₃, are AFM materials when cooled down below their respective Néel temperatures. We used BMS and Raman spectroscopy to measure the acoustic and optical phonon frequencies at RT. We found a large variation, over ~28%, in the acoustic phonon group velocities in this group of materials with similar crystal structures. Our data indicate that the width of the acoustic phonon peaks is strongly affected by the optical properties and electronic bandgap. The acoustic phonon lifetime extracted for some of the materials was correlated with their thermal properties. The obtained results are important for understanding the layered antiferromagnetic van der Waals semiconductors and for assessing their potential for optoelectronic and spintronic device applications.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Dylan Wright: Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (lead); Project administration (equal); Validation (equal); Visualization (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Zahra Ebrahim Nataj:** Data curation (equal); Formal analysis (equal); Investigation (equal); Methodology (equal); Validation (equal); Writing – original draft (supporting); Writing – review & editing (supporting). **Erick Guzman:** Data curation (supporting); Investigation (supporting); Methodology (supporting). **Jake Polster:** Data curation (supporting); Investigation (supporting). **Menno Bouman:** Data curation (supporting); Formal analysis (supporting); Methodology (supporting); Validation (supporting). **Fariborz Kargar:** Conceptualization (lead); Data curation (lead); Formal analysis (lead); Funding acquisition (lead); Investigation (lead); Methodology (lead); Project administration (lead); Resources (lead); Supervision (lead); Validation (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead). **Alexander A. Balandin:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (lead); Investigation (equal); Methodology (equal); Project administration (lead); Resources (lead); Supervision (lead); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding authors upon reasonable request.

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