Magnetic and electronic phase transitions probed by nanomechanical resonance

Makars Šiškins,1,∗Martin Lee,1,∗Samuel Mañas-Valero,2 Eugenio Coronado,2
Yaroslav M. Blanter,1 Herre S. J. van der Zant,1 and Peter G. Steeneken1,3,†

1Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628 CJ, Delft, The Netherlands
2Instituto de Ciencia Molecular (ICMol), Universitat de València, c/Catedrático José Beltrán 2, 46980 Paterna, Spain
3Department of Precision and Microsystems Engineering, Delft University of Technology, Mekelweg 2, 2628 CD, Delft, The Netherlands

Two-dimensional (2D) materials enable new types of magnetic and electronic phases mediated by their reduced dimensionality like magic-angle induced phase transitions [1, 2], 2D Ising antiferromagnets [3] and ferromagnetism in 2D atomic layers [4, 5] and heterostructures [6]. However, only a few methods are available to study these phase transitions [3, 4, 6–8], which for example is particularly challenging for antiferromagnetic materials [6]. Here, we demonstrate that these phases can be probed by the mechanical motion: the temperature dependent resonance frequency and quality factor of multilayer 2D material membranes show clear anomalies near the phase transition temperature, which are correlated to anomalies in the specific heat of the materials. The observed coupling of mechanical degrees of freedom to magnetic and electronic order is attributed to thermodynamic relations that are not restricted to van der Waals materials. Nanomechanical resonators, therefore, offer the potential to characterize phase transitions in a wide variety of materials, including those that are antiferromagnetic, insulating or so thin that conventional bulk characterization methods become unsuitable.

A universal method to characterize phase transitions in bulk crystals is via anomalies in the specific heat, that are present at the transition temperature according to Landau’s theory [9]. However, specific heat is difficult to measure in thin micron-sized samples with a mass of less than a picogram [10, 11]. Although coupling between mechanical and electronic/magnetic degrees of freedom might not seem obvious, the intuitive picture behind this coupling is that changes in the electronic/magnetic order and entropy in a material are reflected in its specific heat, which in turn results in variations in the thermal expansion coefficient that affect the tension and resonance frequency. As the specific heat near a phase transition is expected to exhibit a discontinuity [9], the temperature dependent resonance frequency of a suspended membrane can thus be used to probe this transition. Here, we use nanomechanical motion to investigate magnetic order in membranes of semiconducting FePS3, NiPS3 and insulating MnPS3 - antiferromagnetic members of the transition-metal phosphor trisulphides (MPS3) [12], and subsequently discuss results on metallic 2H-TaS2, which exhibits a transition to a charge density wave state [13].

FePS3 is an Ising-type antiferromagnet with a Néel temperature in bulk in the range of \( T_N \approx 118 – 123 \) K [3, 12, 14], exhibiting a distinct feature in its specific heat near \( T_N \) [14]. Ionic layers in FePS3 are stacked in van der Waals planes, that can be exfoliated to thin the crystal down with atomic precision [3]. Using mechanical exfoliation and all-dry viscoelastic stamping [15], we transfer thin flakes of FePS3 over circular cavities etched in an oxidised Si wafer, to form membranes (see the inset in Fig. 1a). Suspended FePS3 devices with thicknesses ranging from 8 to 45 nm are placed in a cryostat and cooled down to a temperature of 4 K. The resonance frequency of the nanodrums is then characterized using a laser interferometry technique [16] (see Fig. 1a and Methods).

The resonance frequency of the fundamental membrane mode, \( f_0(T) \), is measured in the temperature range from 4 to 200 K. Typical resonances are shown in Fig. 1b-d in the antiferromagnetic phase (80 K), near the transition (114 K) and in the paramagnetic phase (132 K), respectively. Figure 2a shows \( f_0(T) \) of the same FePS3 membrane (solid blue curve). Near the phase transition, significant changes in amplitude, resonance frequency and quality factor are observed. To analyze the data further, we first analyze the relation between \( f_0 \) and the specific heat. The decrease in resonance frequency with increasing temperature in Fig. 2a is indicative of a reduction in strain due to thermal expansion of the membrane. The observed changes can be understood by considering the resonance frequency of a bi-axially tensile strained circular membrane:

\[
f_0(T) = \frac{2.4048}{\pi d} \sqrt{\frac{E}{\rho (1 - \nu)}} \epsilon(T),
\]

where \( E \) is the Young’s modulus of the material, \( \nu \) its Poisson’s ratio, \( \rho \) its mass density, \( \epsilon(T) \) the strain and \( T \) the temperature. The linear thermal expansion coefficient of the membrane, \( \alpha_L(T) \), and silicon substrate, \( \alpha_{Si}(T) \), are related to the strain in the membrane [17, 18] as \( \frac{\partial \epsilon(T)}{\partial T} \approx -\alpha_L(T) + \alpha_{Si}(T) \), using the approximation \( \alpha_{SiO_2} \ll \alpha_{Si} \) (see Supplementary Section.
FIG. 1. Characterisation of mechanical resonances in a thin antiferromagnetic FePS$_3$ membrane. (a) Laser interferometry setup. Red interferometric detection laser: $\lambda_{\text{red}} = 632$ nm. Blue actuation laser diode: $\lambda_{\text{blue}} = 405$ nm. VNA - Vector Network Analyzer, CM - Cold Mirror, PBS - Polarizing Beam Splitter, PD - Photodiode, LD - Laser Diode. Inset: optical image of a FePS$_3$ membrane, including electrodes introducing an option for electrostatic control of strain in the membrane. Flake thickness: 45 $\pm$ 0.6 nm; membrane diameter: $d = 10 \mu$m. Scale bar: 30 $\mu$m. (b-d) Amplitude ($A$) and phase ($\phi$) of the fundamental resonance at three different temperatures for the device shown in (a). Filled dots - measured data; solid lines - fit.

1). By combining the given expression for $\frac{d\epsilon(T)}{dT}$ with equation (1) and by using the thermodynamic relation $\alpha_L(T) = \gamma c_v(T)/(3KV_M)$ [19] between $\alpha_L(T)$ and the specific heat (molar heat capacity) at constant volume, $c_v(T)$, we obtain:

$$c_v(T) = 3\alpha_L(T) \frac{KV_M}{\gamma} = 3 \left( \alpha_{\text{Si}} - \frac{1}{\mu^2} \frac{d[j_0(T)]}{dT} \right) \frac{KV_M}{\gamma}. \quad (2)$$

Here, $K$ is the bulk modulus, $\gamma$ the Grüneisen parameter, $V_M = M/\rho$ the molar volume of the membrane and $\mu = \frac{2.4048}{\pi^2} \sqrt{\frac{E}{\rho(1-\nu^2)}}$, that are assumed to be only weakly temperature dependent. The small effect of non-constant volume ($\nu \neq 0.5$) on $c_v$ is neglected.

We use the equation (2) to analyze $f_0(T)$ and compare it to the calculated specific heat for FePS$_3$ from literature [14]. In doing so, we estimate the Grüneisen parameter following the Belomestnykh–Tesleva relation $\gamma \approx \frac{3}{2} \left( \frac{1+\nu}{2-\nu} \right)$ [19, 20]. This is an approximation to Leont’ev’s formula [21], which is a good estimation of $\gamma$ for bulk isotropic crystalline solids within $\sim 10\%$ of uncertainty [19]. Furthermore, we use literature values for the elastic parameters of FePS$_3$ as obtained from first-principles theoretical calculations [22] to derive $E = 103$ GPa, $\nu = 0.304$ and $\rho = 3375$ kg/m$^3$ (see Supplementary Section 2). In Fig. 2a, the steepest part of the negative slope of $f_0(T)$ (solid blue curve) leads to a large peak in $\frac{d[f_0^2(T)]}{dT}$ (solid magenta curve) near 114 K, the temperature which we define as $T_X$ and indicate by the vertical dashed lines. In Fig. 2b the specific heat curve of FePS$_3$ (blue solid line) as estimated from the data in Fig. 2a and equation (2) is displayed. The results are compared to a theoretical model for the specific heat of FePS$_3$ (magenta solid line in Fig. 2b), which is the sum of a phononic contribution from the Debye model (magenta dashed line) and a magnetic contribution as calculated by Takano, et al. [14]. It is noted that other, e.g. electronic contributions to $c_v(T)$ are small and can be neglected in this comparison, as is supported by experiments on the specific heat in bulk FePS$_3$ crystals [14]. The close correspondence in Fig. 2b between the experimental and theoretical data for $c_v(T)$ supports the applicability of equation (2). It also indicates that changes in the Youngs modulus near the phase transition, that can be of the order of a couple of percent [25], are insignificant and that it is the anomaly in $c_v$ of FePS$_3$ which produces the observed changes in resonance frequency and the large peak in $\frac{d[f_0^2]}{dT}$ visible in Fig. 2a.

The abrupt change in $c_v(T)$ of the membrane can be understood from Landau’s theory of phase transitions [9]. To illustrate this, we consider a simplified model for an antiferromagnetic system, like FePS$_3$, with free energy,
$F$, which includes a strain-dependent magnetostriction contribution (see Supplementary Section 3). Near the transition temperature and in the absence of a magnetic field it holds that:

$$F = F_0 + \left[ a(T - T_N) + \zeta(\epsilon) \right] L_z^2 + BL_z^4. \quad (3)$$

Here, $a$ and $B$ are phenomenological positive constants, $L_z$ is the order parameter in the out-of-plane direction and $\zeta(\epsilon) = \eta_{ij} \epsilon_{ij}$, a strain-dependent parameter with $\eta_{ij}$ a material-dependent tensor, that includes the strain and distance dependent magnetic exchange interactions between neighbouring magnetic moments. By minimizing equation (3) with respect to $L_z$, the equilibrium free energy, $F_{\text{min}}$, and order parameter are obtained (see Supplementary Section 3). Two important observations can be made. Firstly, strain shifts the transition temperature according to:

$$T_N(\epsilon) = T_N - \frac{\zeta(\epsilon)}{a}, \quad (4)$$

where $T_N$ is the Neél temperature, below which free energy minima $F_{\text{min}}$ with finite order ($L_z \neq 0$) appear. Secondly, since close to the transition the specific heat follows $c_v(T) = -T \frac{\partial F_{\text{min}}}{\partial T}$, this general model predicts a discontinuity in $c_v$ of magnitude $T_N^2 \frac{\alpha^2}{2B}$ at the transition temperature $T_N^*$, in accordance with the experimental jump in $c_v(T)$ and $\frac{\partial (f_0^2(T))}{\partial T}$ observed in Fig. 2a and b.

We now analyze the quality factor data shown in Fig. 2c,d. Just above $T_N$, the quality factor of the resonance (Fig. 2c) shows a significant increase as the temperature is increased from 114 to 140 K. The observed minimum in the quality factor near the phase transition, suggests that dissipation in the material is linked to the thermodynamics and can be related to thermoelastic damping. We model the thermoelastic damping according to Zener [23] and Lifshitz-Roukes [24] that report dissipation of the form $Q^{-1}(T) = \beta c_v(T) T$, where $\beta$ is the thermomechanical term (see Supplementary Section 4). Since we have obtained an estimate of $c_v(T)$ from the resonance frequency analysis (Fig. 2b), we use this relation to compare the experimental dissipation $Q^{-1}(T)$ (orange solid line) to a curve proportional to $c_v(T) T$ (blue solid line) in Fig. 2d. Both the measured dissipation and the thermoelastic term display a peak near $T_N \sim 114$ K. The close qualitative correspondence between the two quantities is an indication that the thermoelastic damp-
ing related term indeed can account for the temperature dependence of $Q(T)$ near the phase transition. We note that the temperature dependent dissipation in thin membranes is still not well understood, and that more intricate effects might play a role in the observed temperature dependence.

Equation (4) predicts that the transition temperature is strain-dependent due to the distance dependent interaction coefficient $\zeta(\varepsilon)$ between magnetic moments. To verify this effect, we use an $8 \pm 0.5$ nm thin sample of FePS$_3$ suspended over a cavity of $4 \mu$m in diameter. A gate voltage $V_{G}^{DC}$ is applied between the flake and the doped bottom Si substrate to introduce an electrostatic force that pulls the membrane down and thus strains it (see Supplementary Section 5). As shown in Fig. 3a, the resonance frequency of the membrane follows a W-shaped curve as a function of gate voltage. This is due to two counteracting effects [26]; at small gate voltages capacitive softening of the membrane occurs, while at higher voltages the membrane tension increases due to the applied electrostatic force, which causes the resonance frequency to increase.

Figure 3b shows $f_0(T)$ for six different gate voltages. The shift of the point of steepest slope of $f_0(T)$ with increasing $V_{G}^{DC}$ is well visible in Fig. 3b and even more clear in Fig. 3c, where the peak in $\frac{df_0^2}{dT}$ shifts 6 K downward by electrostatic force induced strain. The observed reduction in $T_N$ as determined by the peak position in $\frac{df_0^2}{dT}$ qualitatively agrees with the presented model and its strain dependence from equation (4), as shown in Fig. 3d indicative of a reduced coupling of magnetic moments with increasing distance between them due to tensile strain.

Since the coupling between specific heat and the order parameter in materials is of a general thermodynamic nature, the presented methodology is applicable to a wide variety of materials provided that elastic properties of the material and Gr"uneisen parameter are weakly temperature dependent, the substrate satisfies the condition $\alpha_{\text{sub}} \ll \alpha_{\text{material}}$ and that the frequency shifts and changes in $Q$ are large enough to be resolved. We further demonstrate the method by detecting magnetic phase transitions in NiPS$_3$ and MnPS$_3$. Compared to FePS$_3$, the effect of the phase transitions in MnPS$_3$ and NiPS$_3$ on the resonances is more gradual (see Supplementary Section 2) with both materials showing broader maxima in $\frac{df_0^2(T)}{dT}$ near their $T_N$ at 76 K and 151 K, respectively, which is consistent with measurements of bulk crystals [12, 14].

In order to demonstrate the detection of an electronic
In conclusion, we have demonstrated a method for identifying phase transitions in ultrathin membranes of 2D materials via their mechanical resonance. An analytical equation for the relation between the specific heat of the material and the temperature dependent resonance frequency is derived and shown to be in good agreement with experimental results. The presented methodology thus shows that mechanical motion of suspended membranes can be used to probe magnetic and electronic order in membranes. Since the materials are characterised in a suspended state, substrate effects on the electronic and magnetic properties of the thin materials are excluded. The technique is particularly appealing for the characterisation of ultrathin membranes of antiferromagnetic and insulating materials that are difficult to characterize otherwise. It is anticipated that it can be applied to a large range of van der Waals materials [6, 8], thin 2D complex oxide sheets [28, 29] and organic antiferromagnets [30], contributing to a better understanding of fundamental models of magnetism and other ordering mechanisms in two dimensions.

**METHODS**

**Sample fabrication** To realize electrical contact to the samples for electrostatic experiments, Ti/Au electrodes are pre-patterned by a lift-off technique. Cavities are defined by reactive ion etching of circular holes with a diameter of $4 \times 10^{-4}$ mm in oxidized doped silicon wafers with an SiO$_2$ thickness of 285 nm. Flakes of van der Waals crystals are exfoliated from high quality synthetically grown crystals with known stoichiometry (see Supplementary Section 7). All flakes are transferred on a pre-patterned chip by an all-dry viscoelastic stamping directly after exfoliation. Subsequently, samples are kept in an oxygen free environment to avoid degradation.

**Controlled measurement environment** The samples are mounted on a piezo-based $xy$ nanopositioning stage inside a chamber of a closed-cycle cryostat with optical access. A closed feedback loop controlled local sample heater is used to perform temperature sweeps at a rate of $\sim 5 \, \text{K/min}$, while keeping the pressure in the chamber below $10^{-9}$ mbar. During the data acquisition temperature is kept constant with $\sim 10 \, \text{mK}$ stability.

**Laser interferometry** A blue diode laser ($\lambda = 405$ nm), which is power-modulated by a Vector Network Analyzer (VNA), is used to excite the membrane and optothermally drive it into motion. Displacements are detected by focusing a red He-Ne laser beam ($\lambda = 632$ nm) on the cavity formed by the membrane and Si substrate. The reflected light, which is modulated by the position-dependent membrane motion, is recorded by a photodiode and processed by a phase-sensitive VNA. All measurements are performed at incident laser powers of $P_{\text{red}} < 10 \, \mu\text{W}$ and $P_{\text{blue}} < 0.6 \, \mu\text{W}$. It is checked for all membranes that the resonance frequency changes due to laser heating are insignificant. Laser spot size is on the
order of $\sim 1 \mu m$. The uncertainty in measured transition temperatures is estimated from determining the peak position in $-\frac{dI(T)}{dT}$ within 2% accuracy in the measured maximum.

**Atomic Force Microscopy** AFM inspections to determine sample thickness are performed in tapping mode on a Bruker Dimension FastScan AFM. We use cantilevers with spring constants of $k = 30 - 40$ N/m. Error bars on reported thickness values are determined by measuring three to five profile scans of the same flake.

**ACKNOWLEDGMENTS**

M.Š., M.L., H.S.J.v.d.Z. and P.G.S. acknowledge funding from the European Union’s Horizon 2020 research and innovation program under grant agreement number 785219. H.S.J.v.d.Z., E.C. and S.M.-V. thank COST Action MP1607.

[1] Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. *Nature* **556**, 43–50 (2018).

[2] Sharpe, A. L. et al. Emergent ferromagnetism near three-quarters filling in twisted bilayer graphene. *Science* **365**, 605–608 (2019).

[3] Lee, J.-U. et al. Ising-type magnetic ordering in atomically thin FePS$_3$. *Nano Lett.* **16**, 7433–7438 (2016).

[4] Huang, B. et al. Layer-dependent ferromagnetism in a van der Waals crystal down to the monolayer limit. *Nature* **546**, 270–273 (2017).

[5] Gong, C. et al. Discovery of intrinsic ferromagnetism in two-dimensional van der Waals crystals. *Nature* **546**, 265–269 (2017).

[6] Gibertini, M., Koperski, M., Morpurgo, A. F. & Novoselov, K. S. Magnetic 2D materials and heterostructures. *Nat. Nanotechnol.* **14**, 408–419 (2019).

[7] McGuire, M. A., Dixit, H., Cooper, V. R. & Sales, B. C. Coupling of crystal structure and magnetism in the layered, ferromagnetic insulator CrI$_3$. *Chem. Mater.* **27**, 612–620 (2015).

[8] Novoselov, K. S., Mishchenko, A., Carvalho, A. & Neto, A. H. C. 2D materials and van der Waals heterostructures. *Science* **353**, aac9439 (2016).

[9] Landau, L. D., Pitaevskii, L. P. & Lifshitz, E. M. *Electrodynamics of continuous media*, vol. 8 (Butterworth, New York, 1984), 2 edn.

[10] Morell, N. et al. Optomechanical measurement of thermal transport in two-dimensional MoSe$_2$ lattices. *Nano Lett.* **19**, 3143–3150 (2019).

[11] Doleman, R. J. et al. Transient thermal characterization of suspended monolayer MoS$_2$. *Phys. Rev. Mater.* **2**, 114008 (2018).

[12] Joy, P. A. & Vasudevan, S. Magnetism in the layered transition-metal thiophosphates MPS$_3$ (M=Mn, Fe, and Ni). *Phys. Rev. B* **46**, 5425–5433 (1992).

[13] Abdel-Hafiez, M. et al. Enhancement of superconductivity under pressure and the magnetic phase diagram of tantalum disulfide single crystals. *Sci. Rep.* **6** (2016).

[14] Takano, Y. et al. Magnetic properties and specific heat of MPS$_3$ (M=Mn, Fe, Zn). *J. Magn. Magn. Mat.* **272–276**, E593–E595 (2004).

[15] Castellanos-Gomez, A. et al. Deterministic transfer of two-dimensional materials by all-dry viscoelastic stamping. *2D Mater.* **1**, 011002 (2014).

[16] Castellanos-Gomez, A. et al. Single-layer MoS$_2$ mechanical resonators. *Adv. Mater.* **25**, 6719–6723 (2013).

[17] Morell, N. et al. High quality factor mechanical resonators based on WS$_2$ monolayers. *Nano Lett.* **16**, 5102–5108 (2016).

[18] Singh, V. et al. Probing thermal expansion of graphene and modal dispersion at low-temperature using graphene nanoelectromechanical systems resonators. *Nanotechnology* **21**, 165204 (2010).

[19] Sanditov, D. S. & Belomestnykh, V. N. Relation between the parameters of the elasticity theory and averaged bulk modulus of solids. *Tech. Phys.* **56**, 1619–1623 (2011).

[20] Belomestnykh, V. N. & Tesleva, E. P. Interrelation between anharmonicity and lateral strain in quasi-isotropic polycrystalline solids. *Tech. Phys.** 49**, 1098–1100 (2004).

[21] Leon’ev, K. On connection between elastic and thermal properties of substances. *Akusticheskij Zhurnal* **27**, 554–561 (1981).

[22] Hashemi, A., Komsa, H.-P., Puska, M. & Krasheninnikov, A. V. Vibrational properties of metal phosphorus trichalcogenides from first-principles calculations. *J. Phys. Chem. C* **121**, 27207–27217 (2017).

[23] Zener, C. Internal friction in solids I. Theory of internal friction in reeds. *Phys. Rev.* **52**, 230–235 (1937).

[24] Lifshitz, R. & Roukes, M. L. Thermoelastic damping in micro- and nanomechanical systems. *Phys. Rev. B* **61**, 5600–5609 (2000).

[25] Barmatz, M., Testardi, L. R. & Salvo, F. J. D. Elasticity measurements in the layered dichalcogenides TaSe$_2$ and NbSe$_2$. *Phys. Rev. B* **12**, 4367–4376 (1975).
[26] Lee, J. et al. Electrically tunable single- and few-layer MoS$_2$ nanoelectromechanical systems with broad dynamic range. *Sci. Adv.* **4**, eaa6653 (2018).

[27] Saint-Paul, M. & Monceau, P. Survey of the thermo-dynamic properties of the charge density wave systems. *Adv. Condens. Matter Phys.* **2019**, 1–14 (2019).

[28] Ji, D. et al. Freestanding crystalline oxide perovskites down to the monolayer limit. *Nature* **570**, 87–90 (2019).

[29] Davidovikj, D. et al. Ultrathin complex oxide nanomechanical resonators (2019). Preprint at https://arxiv.org/abs/1905.00056v1.

[30] López-Cabrelles, J. et al. Isoreticular two-dimensional magnetic coordination polymers prepared through presynthetic ligand functionalization. *Nat. Chem.* **10**, 1001–1007 (2018).
SUPPLEMENTARY INFORMATION: Magnetic and electronic phase transitions probed by nanomechanical resonance

Makars Víšků,1,∗ Martin Lee,1,∗ Samuel Mañas-Valero,2 Eugenio Coronado,2 Yaroslav M. Blanter,1 Herre S. J. van der Zant,1 and Peter G. Steeneken1,3,†

1 Kavli Institute of Nanoscience, Delft University of Technology, Lorentzweg 1, 2628 CJ, Delft, The Netherlands
2 Instituto de Ciencia Molecular (ICMol), Universitat de València, c/Catedrático José Beltrán 2, 46980 Paterna, Spain
3 Department of Precision and Microsystems Engineering, Delft University of Technology, Mekelweg 2, 2628 CD, Delft, The Netherlands

CONTENTS

1. Fundamental resonance frequency of a circular plate and membrane 1
2. Mechanical resonances and specific heat of MPS3 (M=Fe, Ni, Mn) 2
3. Entropy in a suspended antiferromagnet 4
4. Dissipation and thermoelastic damping in vibrating membranes 5
5. Electric field induced strain in a circular FePS3 membrane 6
6. Mechanical resonances and specific heat of 2H-TaS2 near the charge density wave transition 8
7. Crystal growth and characterization 8

References 10

1. FUNDAMENTAL RESONANCE FREQUENCY OF A CIRCULAR PLATE AND MEMBRANE

In this section we analyze in more detail the resonance frequency of the FePS3 resonators and show that near the phase transition they are close to the membrane limit. The fundamental resonance frequency of the mechanical resonator, \( f_0 \), in the crossover membrane-plate regime can be approximated as [1, 2]:

\[
\begin{align*}
  f_0(T) &\approx \sqrt{f_{\text{membrane}}^2 + f_{\text{plate}}^2} = \\
  &\sqrt{\left(\frac{2.4048}{\pi d}\right)^2 \frac{E}{\rho (1-\nu)} + \left(\frac{10.21t}{\pi d^2}\right)^2 \frac{E}{3\rho (1-\nu^2)}},
\end{align*}
\]

(S1)

where \( d \) is the diameter of the membrane, \( E \) the Young’s modulus, \( \epsilon(T) \) the strain, \( \nu \) the Poisson’s ratio, \( t \) the thickness, \( \rho \) the mass density and \( T \) the temperature. The resonance frequency of the fundamental mode of a circular resonator is thickness dependent. For plate resonators \( f_{\text{plate}} \propto t \), as expected for small and linear deflection [1–3]. For membranes, however, \( f_0 \) is dominated by the biaxial tension \( N \):

\[
f_{\text{membrane}} = \frac{2.4048}{\pi d} \sqrt{\frac{N}{\rho t}}.
\]

(S2)

∗ These authors contributed equally.
† e-mail: p.g.steeneken@tudelft.nl; h.s.j.vanderzant@tudelft.nl; m.víšků-1@tudelft.nl
Equation (S2) yields $f_{\text{membrane}} \propto t^{-0.5}$ for thin resonators. When the membrane is subjected to temperature changes, the total tension is dominated by thermal strains $\epsilon_t^h$. Thermal strains are of dilatational nature and do not cause any shear, thus, these can be written as: $\epsilon_t^h = \alpha \Delta T$, where $\alpha$ is the thermal expansion coefficient. In-plane radial thermal strain is then related to tension according to the Hooke’s law as $N = N_0 + E t \epsilon_t^h / (1 - \nu)$, where $N_0$ the intrinsic pre-tension introduced during the fabrication process. Thus, equation (S1) is used to determine if the membranes under study are in the plate or in the membrane limit at a given temperature, as shown in Fig. S1a,b.

The relative contribution of the plate term to the frequency of the resonator is largest at room temperature because the membrane tension is the lowest. As shown in Fig. S1a at $T = 300 \text{ K}$ and $N_0 = 0.1 \text{ N/m}$, thicker FePS$_3$ samples ($t > 40 \text{ nm}$) behave as circular plates. However, as shown in Fig. S1b, in proximity of the transition temperature ($T \approx 114 \text{ K}$) due to temperature-induced strain $\epsilon(T)$ (see equation (S1)), the resonator behaves close to the membrane limit over a thickness range from zero to 60 nm.

The total strain in the membrane is estimated using $\epsilon(T) = \epsilon_0 - \int_{300 \text{ K}}^T (\alpha_{\text{material}}(T) - \alpha_{\text{Si}}(T))dT$, where $\epsilon_0$ is the intrinsic pre-strain at $T = 300 \text{ K}$. Because $\alpha_{\text{SiO}_2} \ll \alpha_{\text{Si}}$ [4, 5], the effect of the thin SiO$_2$ layer can be neglected. As shown in Fig. S1c, the thermal expansion coefficient of the silicon substrate (solid magenta line) is small compared to that of FePS$_3$. Therefore, the total strain in the membrane will mainly build up due to $\alpha_{\text{FePS}_3}$, and this term dominates the change in resonance frequency, $f_0(T)$ as depicted in Fig. S1d.

2. MECHANICAL RESONANCES AND SPECIFIC HEAT OF MPS$_3$ (M=Fe, Ni, Mn)

In addition to FePS$_3$, we measure MnPS$_3$ and NiPS$_3$ membranes with a 10 $\mu$m diameter and thicknesses of 31.8±1.2 and 35.7±1.1 nm, respectively. Comparative study of these is particularly interesting since FePS$_3$ is an Ising antiferromagnet, while the other two are Heisenberg (MnPS$_3$) and XY (NiPS$_3$) antiferromagnets. The resonance peak of the fundamental membrane mode, $f_0(T)$, as well as the Q-factor is measured from 4 – 200 K using the
procedure described in the main text.
At the phase transition, significant changes in both the resonance frequency (Fig. S2a) and quality factor (Fig. S2b) of

![Graphs](image)

FIG. S2. Mechanical properties of MnPS₃ and NiPS₃ membranes. Dashed vertical lines indicate transition temperatures, Tₙ.
(a) Solid blue line - Measured resonance frequency of MnPS₃ membrane as a function of temperature. Solid magenta line - Temperature derivative of f₀. (b) Measured mechanical damping (Q⁻¹) as a function temperature. Inset: Optical image of the sample, t = 32.3 ± 0.4 nm. Scale bar: 40 μm. (c) Solid blue line - Measured resonance frequency of NiPS₃ membrane as a function of temperature. Solid magenta line - Temperature derivative of f₀. (b) Measured mechanical damping (Q⁻¹) as a function temperature. Inset: Optical image of the sample, t = 35.7 ± 0.5 nm. Scale bar: 30 μm.

the MnPS₃ membrane are observed. Figure S2a shows the resonance frequency (solid blue line) and the corresponding \( \frac{df_0}{dT} \) (solid magenta line) with a peak that occurs at a temperature similar to the transition temperature from the antiferromagnetic phase (< 78 K) to the paramagnetic phase (> 78 K) in the bulk material [6] and is thus attributed as Tₙ. The temperature dependence of \( \frac{df_0}{dT} \) shows a broad hump with a smeared peak at Tₙ, which resembles the temperature dependent specific heat \( c_v(T) \) of this material in bulk form [6]. Figure S2b shows the mechanical dissipation, Q⁻¹, that exhibits a local minimum close to Tₙ as well as a local maximum at \( T \sim 36 \) K. In Fig. S2c, the resonance frequency of the NiPS₃ membrane (solid blue line) is shown with the corresponding \( \frac{df_0}{dT} \) (solid magenta line). A small peak is noticeable in \( \frac{df_0}{dT} \) near bulk \( T_\text{N} \sim 155 \) K indicating the phase transition [7]. However, as shown in Fig. S2d, no significant anomalies in the Q-factor were observed in the case of NiPS₃. Compared to FePS₃, the effect of the phase transitions in MnPS₃ and NiPS₃ on the resonances is more gradual (Fig. S2a-d). Both materials show a peak in \( \frac{df_0}{dT} \) at the Tₙ, but their dissipation does not show a clear jump at Tₙ like in the case of FePS₃.

We also calculated the corresponding temperature dependent specific heat \( c_v(T) \) for the three MPS₃ samples. Following the methodology described in the main text, we estimate the Grüneisen parameter following the Belomestnykh–Tesleva relation [8, 9]: \( \gamma \approx \frac{3}{2} \left( \frac{1+\nu}{2-3\nu} \right) \). We use reported values for monolayers of FePS₃ (\( C_{11} = 72.7 \)
FIG. S3. Estimated specific heat (c_v) for (a) MPS_3 (M=Fe, Mn, Ni) membranes of t = 45.2 ± 0.6 nm, 31.8 ± 1.2 nm and 35.7 ± 1.1 nm, respectively. Dashed lines indicate the corresponding transition temperatures (T_N): T_N = 76 ± 5 K for MnPS_3, T_N = 114 ± 3 K for FePS_3 and T_N = 151 ± 5 K for NiPS_3. (b) Specific heat of ultrathin FePS_3 membrane compared to that of its bulk form. Solid blue line - measured c_v of the FePS_3 ultrathin membrane. Solid magenta line - c_v reported by Takano, et al. [6] for a bulk crystal.

N/m, C_{12} = 22.1 N/m and ρ_{2D} = 2.16×10^{-6} kg/m^2), MnPS_3 (C_{11} = 61.7 N/m, C_{12} = 20.2 N/m and ρ_{2D} = 2.00×10^{-6} kg/m^2) and NiPS_3 (C_{11} = 87.0 N/m, C_{12} = 23.1 N/m and ρ_{2D} = 2.15×10^{-6} kg/m^2) as obtained from first-principles calculations [10, 11]. We convert these to three-dimensional Young’s modulus E, mass density ρ and Poisson’s ratio ν using E = \frac{C_{11}-C_{12}}{C_{12}}, ρ = \frac{E\nu}{(1-\nu^2)} and ν = \frac{C_{12}}{C_{11}}; assuming the interlayer distance for the compounds to be t_{FePS_3} = 0.64 nm, t_{MnPS_3} = 0.65 nm and t_{NiPS_3} = 0.64 nm as determined from Fig. S7a-c. The resulting values are E = 103 GPa, ν = 0.304 and ρ = 3375 kg/m^3 for FePS_3, E = 85 GPa, ν = 0.327 and ρ = 3076 kg/m^3 for MnPS_3 and E = 126 GPa, ν = 0.265 and ρ = 3359 kg/m^3 for NiPS_3.

In Fig. S3a the specific heat for the three MPS_3 are displayed, as determined from the data in Fig. S2a,c, and Fig. 2a using equation (2) from the main text. Detected temperatures are indicated by dashed lines and found to be T_N = 76 ± 5 K for MnPS_3, T_N = 114 ± 3 K for FePS_3 and T_N = 151 ± 5 K for NiPS_3, which are in agreement with experiments in bulk crystals of MPS_3 [6, 12]. As shown in Fig. S3b, for FePS_3 we obtain a good correspondence to bulk literature values [6] (solid magenta curve) without fitting parameters. The transition related peak in c_v, however, is more pronounced in the case of the ultrathin membrane (solid blue curve) than in the bulk.

3. ENTROPY IN A SUSPENDED ANTIFERROMAGNET

A uniaxial antiferromagnet with two antiparallel magnetic sub-lattices close to the phase transition can be modeled by the Landau theory of phase transitions [13, 14]. In this theory, the vector order parameter \textbf{L} is defined as the difference between the magnetizations of the two sublattices, \textbf{M}_1 and \textbf{M}_2 (thus, the antiferromagnetic vector \textbf{L} = \textbf{M}_1 - \textbf{M}_2). This order parameter is zero in the paramagnetic phase and is finite in the antiferromagnetic phase. The magnetization, \textbf{M}, is the sum of both magnetizations (\textbf{M} = \textbf{M}_1 + \textbf{M}_2) and equals to zero in the absence of an external magnetic field, \textbf{H}.

For a uniaxial crystal antiferromagnet, the transition temperature is commonly known as Néel temperature, T_N, but, as conveyed by Landau, et al. [13, 14], it is also referred to as the antiferromagnetic Curie temperature and denoted as T_C. Note that further we will consider the Néel temperature, denoted as T_N, as the temperature where the transition from a paramagnetic to an antiferromagnetic phase takes place. Near T_N, \textbf{L} is small and the free energy, F, can be expanded in terms of \textbf{L} and \textbf{H}, since the magnetization is only non-zero when an external field \textbf{H} is present (i.e. a spin-flip transition). Following Landau formalism and considering z-axis as the main axis of symmetry, we write (see e.g. Ref. 14):

\[ F = F_0 + A L^2 + B L^4 + D (\textbf{H} \cdot \textbf{L})^2 + D' H^2 L^2 - \frac{1}{2} \chi_p H^2 + \frac{1}{2} \beta (L_x^2 + L_y^2) - \frac{1}{2} \mu M (H_x^2 + H_y^2) - \frac{H^2}{8\pi}, \]

where A = a(T - T_N), D, D', a and B are phenomenological positive constants which are taken temperature independent, \chi_p the isotropic susceptibility for T > T_N, \beta the index that describes the temperature dependence of
the spontaneous magnetization below \( T_N \) (\( \beta > 0 \) for \( \mathbf{L} \) directed out-of-plane) and \( \mu_M \) the magnetic susceptibility in the paramagnetic phase. The minimization of equation (S3), where the vector \( \mathbf{L} \) is along the \( z \)-axis (thus, \( L_x = L_y = 0 \) and \( \beta > 0 \)) and in the absence of field (\( H = 0 \)), gives \( L_x = L_y = 0 \) and \( L_z = \sqrt{a(T_N - T)/(2B)} \) for \( T > T_N \) (paramagnetic phase) and \( L_z = \sqrt{a(T_N - T)/(2B)} \) for \( T < T_N \) (antiferromagnetic phase).

Now we introduce strain. For an easy-axis antiferromagnet near \( T_N \) with the vector \( \mathbf{L} \) along the \( z \)-axis and in the absence of field, we can write equation (S3) as:

\[
F = F_0 + A L^2 + B L^4 + \frac{1}{2} \beta (L_x^2 + L_y^2) + \zeta L_z^2 + \zeta_x L_x^2 + \zeta_y L_y^2,
\]

(S4)

where the last three added terms describe the magnetostriiction effects, i.e., the coupling of magnetic moments to strain; the coefficients \( \zeta, \zeta_{x,y} \) are linear combinations of the components of the strain tensor. We assume that the strain is determined by the deformation of the membrane, and that the back-action exerted by the magnetization on the strain is negligible. In that case, for the calculation of the order parameter, \( \zeta \) and \( \zeta_{x,y} \) can be treated as temperature-independent constants. We also assume that \(|\zeta|, |\zeta_{x,y}| \ll \beta\), so that even the strained antiferromagnet exhibits an easy-axis. The minimization of equation (S4) gives \( L_x = L_y = 0 \), leading to a free energy which only depends on \( L_z \),

\[
F = F_0 + |a(T - T_N) + \zeta|L_z^2 + BL_z^4.
\]

(S5)

The first observation is that the magnetostriction effects shift the antiferromagnetic phase transition point. Indeed, the phase transition occurs at the temperature \( T_N^* \) at which the coefficient multiplying \( L_z^2 \) in equation (S5) vanishes. This gives \( T_N^* = T_N - \zeta/a \) (which is equation (4) in the main text).

Second, we calculate the behavior of the specific heat close to the phase transition. Minimizing equation (S5) with respect to \( L_z \), we find the equilibrium free energy, \( F_{\text{min}} = F_0 - a^2(T - T_N^*)^2/(2B) \) in the antiferromagnetic phase, where \( F_0 \) is the free energy of the paramagnetic phase. We proceed by calculating the entropy \( S_{\text{min}} = -\partial F_{\text{min}}/\partial T \),

\[
S_{\text{min}} - S_0 = \begin{cases} -a^2(T_N^* - T)/(2B) & \text{for } T < T_N^* \\ 0 & \text{for } T > T_N^* \end{cases},
\]

(S6)

where \( S_0 = -\partial F_0/\partial T \) is the entropy of the paramagnet with \( S_0 \) the non-magnetic contribution to the entropy. Since the renormalized transition temperature \( T_N^* \) is strain-dependent, the entropy of the antiferromagnet contains an additional (as compared to the paramagnetic phase) strain-dependent term. The specific heat near \( T_N^* \), \( c_{v,\text{min}} = T\partial S_{\text{min}}/\partial T \), reads

\[
c_{v,\text{min}} - c_{v,0} = \begin{cases} T_N^* a^2/(2B) & \text{for } T < T_N^* \\ 0 & \text{for } T > T_N^* \end{cases},
\]

(S7)

where \( c_{v,0} \) is the non-magnetic contribution to the specific heat. This derivation shows, in line with Landau theory [13, 14], that the specific heat has a jump at the transition temperature, \( T_N^* \).

4. DISSIPATION AND THERMOELASTIC DAMPING IN VIBRATING MEMBRANES

For a membrane in motion the dissipation \( Q^{-1} \) is defined as the ratio of the energy lost per cycle to \( 2\pi \) times the stored energy. The total dissipation in a membrane is given by the sum of all contributing dissipation mechanisms [15]:

\[
Q^{-1} = Q_{\text{medium}}^{-1} + Q_{\text{damping}}^{-1} + Q_{\text{intrinsic}}^{-1} + Q_{\text{other}}^{-1}.
\]

(S8)

\( Q_{\text{medium}}^{-1} \) is related to losses due to the interaction with a fluid medium or gas and can thus be neglected in high vacuum. \( Q_{\text{damping}}^{-1} \) is related to the transfer of mechanical energy to the anchoring substrate, which has a small temperature dependence [15]. \( Q_{\text{intrinsic}}^{-1} \) quantifies all intrinsic damping mechanisms of the material, such as thermoelastic damping, internal and surface friction, and phonon-phonon interaction loss. \( Q_{\text{other}}^{-1} \) relates all other possible damping mechanisms, such as electrical charge damping and magneto-motive damping. In this section we focus on deriving the expression for the thermoelastic damping. Further derivation in this section follows Zener’s standard linear solid model [16, 17].

The dissipation is equal to the ratio between the imaginary and real parts of the complex elastic modulus \( E^*(\omega) = E'(\omega) + iE''(\omega) \):

\[
Q^{-1} = \frac{E''}{E'}.
\]

(S9)
For a standard linear solid with a single relaxation mechanism, real and imaginary parts of the complex elastic modulus are given by:

\[ E'(\omega) = E_r + (E_u - E_r) \frac{\omega^2 \tau^2}{1 + (\omega \tau)^2}, \]
\[ E''(\omega) = (E_u - E_r) \frac{\omega \tau}{1 + (\omega \tau)^2}, \] (S10)

where \( \omega \) is the resonance eigenfrequency, \( \tau \) the thermal relaxation time, \( E_r \) and \( E_u \) are the relaxed (or isothermal) and unrelaxed (or adiabatic) Young’s moduli, respectively (see Ref. 15 and 17 for more details). From equation (S9), the dissipation is then:

\[ Q^{-1} = \frac{(E_u - E_r) \frac{\omega \tau}{E_r + E_u(\omega \tau)^2}}{E_r + E_u(\omega \tau)^2} \approx \frac{E_u - E_r}{E_r} \frac{\omega \tau}{1 + (\omega \tau)^2}, \] (S11)

for \((E_u - E_r) \ll E_r \approx E_u\). For a standard thermoelastic solid and in the case of thermoelastic damping \( Q_{TED}^{-1} \), equation (S11) can be rewritten as [16–18]:

\[ Q_{TED}^{-1} = \frac{E_u - E_r}{E_r} \beta = \frac{E \alpha^2 T}{c_v} \beta, \] (S12)

where \( \alpha \) is the thermal expansion coefficient, \( c_v \) the specific heat and \( \beta \) the thermomechanical parameter, that in Zener’s model is \( \beta_Z = \frac{\omega \tau}{1 + (\omega \tau)^2} \). The exact expression for thermoelastic damping and its relation to the thermal properties of solids was found by Lifshitz and Roukes [18] with \( \beta_{LR} = \frac{6}{\xi^2} - \frac{6}{\xi^2} \frac{\sinh(\xi) + \sin(\xi)}{\cosh(\xi) + \cos(\xi)} \), where \( \xi = \frac{\pi}{\sqrt{2}} \sqrt{\omega \tau} \). Under the assumption that the temperature dependence of \( \omega \tau \) is small, equation (S12) can be written as:

\[ Q_{TED}^{-1} \propto \frac{E \alpha^2 T}{c_v}. \] (S13)

The close correspondence between this expression and the data in Fig. 2d and 4d in the main text indicates that this assumption is reasonable.

As could be noted from equation (2) in the main text and assuming the elastic properties of the material and its Grüneisen parameter to have a negligible temperature dependence at low temperatures, the thermoelastic damping \( Q_{TED}^{-1} \) is related to the frequency \( f_0 \) of the resonator as:

\[ Q_{TED}^{-1}(T) \propto \frac{E \alpha^2(T)}{c_v(T)} \propto \frac{c_v(T)}{T} \propto -T \frac{d[f_0^2(T)]}{dT}, \] (S14)

where we use \( \alpha(T) \propto c_v(T) \) and \( c_v(T) \propto \frac{d[f_0^2(T)]}{dT} \) according to equation (2) in the main text. Therefore, in accordance with our observations in the main text, an anomaly (a jump) in the specific heat at the transition temperature will be visible in both \( \frac{d[f_0^2(T)]}{dT} \) and \( Q^{-1}(T) \) if the thermoelastic damping is the dominating dissipation mechanism.

5. ELECTRIC FIELD INDUCED STRAIN IN A CIRCULAR FePS₃ MEMBRANE

A constant electrostatic load is applied to the circular membrane with a radius \( a \) (see Fig. S4). This results in a uniform curvature \( R \) with a maximum deflection \( \delta \).

![Image of a membrane deformation](image)

**FIG. S4.** Schematic of the membrane deformation, cross-section view.

In this system, the electrostatic load is balanced by the total tension \( N_{total} \):

\[ \frac{\varepsilon_0 (V_{DC}^2)}{2(g_0 - \delta)^2} \pi R^2 = 2\pi N_{total} R, \] (S15)
where $\varepsilon_0$ is the vacuum permittivity, $V_{DC}^G$ the applied voltage, $g_0$ the gap size between the membrane and the bottom silicon plate. For small deflections the radius of curvature can be approximated as:

$$R \approx \frac{a^2}{2\delta}.$$  \hspace{1cm} (S16)

so that the tension $N_{\text{total}}$ becomes:

$$N_{\text{total}} = \frac{\varepsilon_0 (V_{DC}^G)^2}{2(g_0 - \delta)^2} \frac{a^2}{4\delta}.$$  \hspace{1cm} (S17)

The radial strain in such a membrane can be estimated from the arc length [19, 20]:

$$\epsilon = \frac{R\theta - a}{a} \approx \frac{a^2}{6R^2}.$$  \hspace{1cm} (S18)

Combining this result with equation (S16) yields:

$$\epsilon \approx \frac{2\delta^2}{3a^2}.$$  \hspace{1cm} (S19)

From Hooke’s law and including equation (S19), one can write the strain due to deformation as:

$$N = \frac{Et}{1 - \nu} \epsilon = \frac{2Et\delta^2}{3a^2(1 - \nu)},$$  \hspace{1cm} (S20)

Therefore, the total tension in the membrane, including the thermal expansion induced tension $N_0(T)$ at a certain temperature $T$, can be written as:

$$N_{\text{total}} = N_0(T) + N = N_0(T) + \frac{2Et\delta^2}{3a^2(1 - \nu)} = \frac{\varepsilon_0 (V_{DC}^G)^2}{2(g_0 - \delta)^2} \frac{a^2}{4\delta},$$  \hspace{1cm} (S21)

which also can be rewritten as:

$$V_{DC}^G = \sqrt{\frac{2(g_0 - \delta)^2}{\varepsilon_0}} \left( \frac{4\delta N_0(T)}{a^2} + \frac{8Et\delta^3}{3a^4(1 - \nu)} \right).$$  \hspace{1cm} (S22)

As shown in Fig. S5a, equation (S22) in combination with (S19) and (S1) fits the experimental data well and is used to provide an an estimate of the electrostatically induced strain in the membrane (see Fig. S5b).
6. MECHANICAL RESONANCES AND SPECIFIC HEAT OF 2H-TaS$_2$ NEAR THE CHARGE DENSITY WAVE TRANSITION

For 2H-TaS$_2$ resonance frequency measurements were performed on a $d = 4 \mu$m drum made of a 31 nm thin flake (see Fig. S6a, solid blue line). As shown by the solid green line in Fig. S6a, the specific heat-related temperature derivative of $f_0^2$ reveals a clear peak at $T_{CDW} \sim 75$ K. We convert the measured $\frac{d(f_0^2)}{dT}$ to the specific heat using the methodology described in the main text. We use reported values of $E_{2D} = 87$ N/m and $\nu = 0.27$ for a monolayer of 1H-TaS$_2$ obtained from molecular dynamics simulations [21] and mass density $\rho = 6110$ kg/m$^3$. We find the corresponding $E = E_{2D}/t = 149$ GPa, taking the interlayer spacing $t = 0.58$ nm. The estimated specific heat of the 2H-TaS$_2$ membrane is depicted in Fig. S6b (solid blue line). The four-probe resistance was measured on the same flake to confirm the existence of a CDW transition using a conventional electronic based method as shown in Fig. S6c. The expected characteristic kink in the resistance is visible at $\sim 77$ K, consistent with the CDW transition temperature previously reported in 2H-TaS$_2$ [22].

![CDW Normal](image)

FIG. S6. Mechanical properties of a 2H-TaS$_2$ resonator with membrane thickness of 31.2 $\pm$ 0.6 nm. Dashed vertical lines in the panels indicate the transition temperature, $T_{CDW}$. (a) Solid blue line - resonance frequency as a function of temperature. Solid green line - temperature derivative of $f_0^2$. Inset: optical image of the sample. Scale bar: 20 $\mu$m. (b) Solid blue line - estimated specific heat ($c_v$) and thermal expansion coefficient ($\alpha_L$). (c) Solid blue line - four-point resistance of the same sample. Solid green line - $\frac{1}{R} \frac{dR}{dT}$ plot showing the CDW related feature at $T_{CDW}$.

7. CRYSTAL GROWTH AND CHARACTERIZATION

The crystal growth of MPS$_3$ ($M = $ Mn, Fe, Ni) was performed following a typical solid state reaction. Powders of Mn ($> 99.9\%$, from Sigma-Aldrich), Fe ($99.998\%$, from Alfa-Aesar), Ni ($99.99\%$, from Sigma-Aldrich), P ($> 99.99\%$, from Sigma-Aldrich) and S ($99.998\%$, from Sigma-Aldrich) were mixed in a stoichiometric ratio, pressed into a pellet and sealed in an evacuated quartz ampoule ($P \sim 5 \times 10^{-5}$ mbar, length = 25 cm, internal diameter = 1.5 cm) and heated from room temperature to 400 °C at 1.1 °C/min. Then, the temperature was kept constant for twenty days and slowly cooled down (0.07 °C/min).

For obtaining large crystals, 4 mmol of the previous material was mixed with I$_2$ as a transport agent ([I$_2$] $\sim$ 5 mg/cm$^3$) in an evacuated quartz ampoule ($P \sim 5 \times 10^{-5}$ mbar, length = 50 cm, internal diameter = 1.5 cm). The quartz tube was placed inside a three-zone furnace with the material in the leftmost zone. The other two zones were heated up in 24 h from room temperature to 650 °C and kept at that temperature for one day. After this, the leftmost side was heated up to 700 °C in 3 h and a gradient of 700 °C/650 °C/675 °C was established in the three-zone furnace. Then the temperature was kept constant for 28 days and cooled down naturally. As shown in Fig. S7, with this process we could obtain crystals with a length up to several centimeters. The obtained crystals were analyzed by ICP-OES (Inductively Coupled Plasma - Optical Emission Spectrometry) and powder X-ray diffraction. The relative weights of elements obtained are summarized in Table S7. The refinement of the X-ray diffraction pattern (Fig. S7) revealed a monoclinic base-centered crystal system with C12/m1 space group and a unit cell determined by $a = \gamma = 90^\circ$ and $\beta = 107.3(1)^\circ$ (MnPS$_3$), $\beta = 107.13(1)^\circ$ (FePS$_3$), $\beta = 106.945(9)^\circ$ (NiPS$_3$), and $a = 6.077(7)$ Å, $b = 10.55(2)$ Å and $c = 6.805(9)$ Å for MnPS$_3$, $a = 5.939(6)$ Å, $b = 10.296(3)$ Å and $c = 6.716(3)$ Å for FePS$_3$ and $a = 5.815(4)$ Å, $b = 10.087(5)$ Å and $c = 6.627(4)$ Å for NiPS$_3$. The obtained results are in accordance with the ones reported in the literature [23].
FIG. S7. XRPD experimental patterns (black) and corresponding fits (peaks in blue and background in green) for MPS$_3$ (M = Mn, Fe and Ni) crystals. For clarity, the peaks have been marked by red lines. (a) Left: Pattern of MnPS$_3$. Right: Photography of a MnPS$_3$ crystal. The fit gives as a result: $a = 6.077(7)$ Å, $b = 10.55(2)$ Å and $c = 6.805(9)$ Å, $\alpha = \gamma = 90^\circ$ and $\beta = 107.33(1)^\circ$, monoclinic C face center crystal system with C12/m1 space group, $\chi^2 = 5.8 \times 10^{-5}$. (b) Left: Pattern of FePS$_3$. Right: Photography of a FePS$_3$ crystal. The fit gives as a result: $a = 5.939(6)$ Å, $b = 10.296(3)$ Å and $c = 6.716(3)$ Å, $\alpha = \gamma = 90^\circ$ and $\beta = 107.13(1)^\circ$, monoclinic C face center crystal system with C12/m1 space group, $\chi^2 = 1.5 \times 10^{-6}$. (c) Left: Pattern of NiPS$_3$. Right: Photography of a NiPS$_3$ crystal. The fit gives as a result: $a = 5.815(4)$ Å, $b = 10.087(5)$ Å and $c = 6.627(4)$ Å, $\alpha = \gamma = 90^\circ$ and $\beta = 106.945(9)^\circ$, monoclinic C face center crystal system with C12/m1 space group, $\chi^2 = 1.1 \times 10^{-5}$. 
The crystal growth and characterization of 2H-TaS$_2$ was performed as already reported in earlier works [24, 25].

| Element | Obtained (%) | Expected (%) |
|---------|--------------|--------------|
| MnPS$_3$ | Mn 28.0 ± 1.0 | 30.2 |
|         | P 12 ± 1    | 17.0 |
|         | S 49 ± 2    | 52.8 |
| FePS$_3$ | Fe 30.0 ± 1.0 | 30.5 |
|         | P 15.7 ± 0.5 | 16.9 |
|         | S 51 ± 2    | 52.6 |
| NiPS$_3$ | Ni 30.0 ± 1.0 | 31.6 |
|         | P 15.7 ± 0.5 | 16.6 |
|         | S 53 ± 2    | 51.8 |

TABLE SI. Experimental and expected relative weights analyzed by inductively coupled plasma - optical emission spectrometry (ICP-OES) for the different MPS$_3$ crystals (M = Mn, Fe and Ni).

[1] Wah, T. Vibration of circular plates. *J. Acoust. Soc. Am.* 34, 275–281 (1962).
[2] Castellanos-Gomez, A. et al. Single-layer MoS$_2$ mechanical resonators. *Adv. Mater.* 25, 6719–6723 (2013).
[3] Timoshenko, S., Young, D. H. & Weaver, W. *Vibration Problems in Engineering* (Wiley, New York, 1974), 4 edn.
[4] White, G., Birch, J. & Manghnani, M. H. Thermal properties of sodium silicate glasses at low temperatures. *J. Non-Cryst. Solids* 23, 99–110 (1977).
[5] Lyon, K. G., Salinger, G. L., Swenson, C. A. & White, G. K. Linear thermal expansion measurements on silicon from 6 to 340 K. *J. Appl. Phys.* 48, 865–868 (1977).
[6] Takano, Y. et al. Magnetic properties and specific heat of MPS$_3$ (M=Mn, Fe, Zn). *J. Magn. Magn. Mater.* 272–276, E593–E595 (2004).
[7] Kim, K. et al. Suppression of magnetic ordering in XXZ-type antiferromagnetic monolayer NiPS$_3$. *Nat. Commun.* 10 (2019).
[8] Sanditov, D. S., Mashanov, A. A., Darmaev, M. V., Sanditov, B. D. & Mantatov, V. V. Grüneisen parameter and elastic constants of crystals and vitreous bodies. *Russ. Phys. J.* 52, 221–230 (2009).
[9] Belomestnykh, V. N. & Tesleva, E. P. Interrelation between anharmonicity and lateral strain in quasi-isotropic polycrystalline solids. *Tech. Phys.* 49, 1098–1100 (2004).
[10] Hashemi, A., Komsa, H.-P., Puska, M. & Krasheninnikov, A. V. Vibrational properties of metal phosphorus trichalcogenides from first-principles calculations. *J. Phys. Chem. C* 121, 27207–27217 (2017).
[11] Zhang, X., Zhao, X., Wu, D., Jing, Y. & Zhou, Z. MnPSe$_3$ monolayer: A promising 2D visible-light photohydrolytic catalyst with high carrier mobility. *Adv. Sci.* 3, 1600062 (2016).
[12] Joy, P. A. & Vasudevan, S. Magnetism in the layered transition-metal thio phosphates MPS$_3$ (M=Mn, Fe, and Ni). *Phys. Rev. B* 46, 5425–5433 (1992).
[13] Landau, L. D. On the theory of phase transitions. *Zh. Eksp. Teor. Fiz.* 19, 19–32 (1937). [Ukr. J. Phys. 53, 25 (2008)].
[14] Landau, L. D., Pitaevskii, L. P. & Lifshitz, E. M. *Electrodynamics of continuous media*, vol. 8 (Butterworth, New York, 1984), 2 edn.
[15] Schmid, S., Villanueva, L. G. & Roukes, M. L. *Fundamentals of Nanomechanical Resonators* (Springer International Publishing, 2016).
[16] Zener, C. Internal friction in solids. I. Theory of internal friction in reeds. *Phys. Rev.* 52, 230–235 (1937).
[17] Zener, C. Internal friction in solids II. General theory of thermoelastic internal friction. *Phys. Rev.* 53, 90–99 (1938).
[18] Lifshitz, R. & Roukes, M. L. Thermoelastic damping in micro- and nanomechanical systems. *Phys. Rev. B* 61, 5600–5609 (2000).
[19] Small, M. K. & Nix, W. Analysis of the accuracy of the bulge test in determining the mechanical properties of thin films. *J. Mater. Res. Technol.* 7, 1553–1563 (1992).
[20] Weber, P., Güttinger, J., Tsoutsios, I., Chang, D. E. & Bachtold, A. Coupling graphene mechanical resonators to superconducting microwave cavities. *Nano Lett.* 14, 2854–2860 (2014).
[21] Jiang, J.-W. & Zhou, Y.-P. Parameterization of Stillinger-Weber potential for two-dimensional atomic crystals. In *Handbook of Stillinger-Weber Potential Parameters for Two-Dimensional Atomic Crystals* (IntechOpen, 2017).
[22] Abdel-Hafiez, M. et al. Enhancement of superconductivity under pressure and the magnetic phase diagram of tantalum disulfide single crystals. *Sci. Rep.* 6 (2016).
[23] Ouvrard, G., Brec, R. & Rouxel, J. Structural determination of some MPS$_3$ layered phases (M = Mn, Fe, Co, Ni and Cd). *Mater. Res. Bull.* 20, 1181–1189 (1985).
[24] Pinilla-Cienfuegos, E. et al. Local oxidation nanolithography on metallic transition metal dichalcogenides surfaces. *Appl. Sci.* 6, 250 (2016).
[25] Navarro-Moratalla, E. et al. Enhanced superconductivity in atomically thin TaS$_2$. Nat. Commun. 7 (2016).