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Magnetic oscillations induced by phonons in non-magnetic materials

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An unexpected finding two decades ago demonstrated that Shockley electron states in noble metal surfaces are spin-polarized, forming a circulating spin texture in reciprocal space. The fundamental role played by the spin degree of freedom was then revealed, even for a non-magnetic system, whenever the spin-orbit coupling was present with some strength. Here, we demonstrate that, similarly to electrons in the presence of spin-orbit coupling, the propagating vibrational modes are also accompanied by a well-defined magnetic oscillation even in non-magnetic materials. Although this effect is illustrated by considering a single layer of the WSe_2 dichalcogenide, the phenomenon is completely general and valid for any non-magnetic material with spin-orbit coupling. The emerging phonon-induced magnetic oscillation acts as an additional effective flipping mechanism for the electron spin and its implications in the transport and scattering properties of the material are evident and profound.

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In materials science, the most conventional point of view is to assume that propagating vibrational collective modes (phonons) are not associated with any magnetic property if the material itself is non-magnetic. In a magnetic material, however, it is natural to expect a magnetic oscillation associated to a phonon mode. Phonons are commonly understood as sinusoidal patterns of atomic displacements, which couple to electron states by the scalar potential induced by these atomic displacements. Electrons have a well defined spin-polarization under spin-orbit interaction, but when time reversal symmetry applies the spin polarizations for opposite momenta cancel each other and the material results to be non-magnetic. There is a clear evidence that the lattice thermal conductivity of diamagnetic materials couples to external magnetic fields¹, which in principle might seem contrary to the idea that phonons do not have any associated magnetic property. Here we show that similarly to electrons, phonons are also accompanied by an induced effective magnetic oscillation when spin-orbit coupling is present even for non-magnetic materials.

Results

In a solid with spin-orbit coupling, electron states are described by two-component spinor wave functions for each \mathbf{k} point spanning the Brillouin zone (BZ), $\Psi_{\mathbf{k},i}(\mathbf{r}) = \begin{pmatrix} u_{\mathbf{k},i}^{\uparrow}(\mathbf{r}) \\ u_{\mathbf{k},i}^{\downarrow}(\mathbf{r}) \end{pmatrix} e^{i\mathbf{k}\mathbf{r}}$. Each electron state has an associated spin-polarization defined as the expectation value of the Pauli vector $\mathbf{m}_{\mathbf{k},i}(\mathbf{r}) = \langle \Psi_{\mathbf{k},i} | \boldsymbol{\sigma} | \Psi_{\mathbf{k},i} \rangle$. Time reversed Kramers pairs at \mathbf{k} and $-\mathbf{k}$ have opposite spin-polarizations that exactly cancel out when integrated, which therefore implies no magnetism. However, the electron spin-polarization is a crucial physical magnitude in many non-magnetic systems with spin-orbit coupling, one of the most outstanding being probably its role in the topological properties of matter.

A phonon excitation consists in a sinusoidal displacement of atoms and an induced (almost) static response of the electron gas which tries to weaken or screen out the electric perturbation generated by these displacements. Therefore, the question of whether a phonon perturbation creates a magnetic oscillation could be suitably treated considering a generalized dielectric theory of dimension 4×4 mixing the magnetic and electric components of the potential². An alternative and more transparent way to see whether an overall magnetic property emerges is to consider the effect of the perturbation on each electron spinor wave function and then integrate over the BZ. The key point is that if a phonon is excited with momentum \mathbf{q} it couples differently with the electrons at \mathbf{k} and $-\mathbf{k}$, the result being that the spin-polarization of electrons with time reversed momenta do not cancel each other. Under these conditions the balance of the electron spin-polarizations within the BZ is broken and the BZ integral gives a finite value and, therefore, a net real-space magnetic oscillation with the same wave number as the phonon \mathbf{q} .

Let us focus on a single frozen phonon-mode (ν) with momentum \mathbf{q} , which produces a perturbation on both components of the periodic part of the electron spinor wave functions, $\delta^{q,\nu} u_{\mathbf{k},i}^{\sigma}(\mathbf{r})$, where $\delta^{q,\nu}$ denotes the self-consistent variation in the context of density-functional perturbation theory^{3,4} and $u_{\mathbf{k},i}^{\sigma}(\mathbf{r})$ is the periodic part of each spinor component. The unit cell periodic part of the amplitude of the frozen charge-spin density wave is obtained by integrating the contributions from all occupied electron states (see Supplementary Note 1)

$$\delta \tilde{n}_{\mathbf{q},\nu}^{\sigma,\sigma'}(\mathbf{r}) = \sum_{\mathbf{k},i}^{\text{occ}} \left\langle u_{\mathbf{k},i}^{\sigma'}(\mathbf{r}) \middle| \delta^{q,\nu} u_{\mathbf{k},i}^{\sigma}(\mathbf{r}) \right\rangle. \quad (1)$$

Therefore, $\delta \tilde{n}_{\mathbf{q},\nu}^{\sigma,\sigma'}(\mathbf{r}) e^{i\mathbf{q}\mathbf{r}}$ would represent the complete oscillation wave in real space. The absence of a magnetic component accompanying a phonon mode would require the off-diagonal elements ($\sigma \neq \sigma'$) to be zero and that both diagonal components are equal to each other, which, in general, are conditions only fulfilled at the Γ point ($\mathbf{q} = \mathbf{0}$). Writing the 2×2 charge-spin matrix of Eq. (1) in terms of Pauli matrices allows to explicitly distinguish the electronic charge, $\delta \tilde{n}_{\mathbf{q},\nu}^0(\mathbf{r})$, and the magnetic components, $\delta \tilde{\mathbf{M}}_{\mathbf{q},\nu}(\mathbf{r}) = (\delta \tilde{n}_{\mathbf{q},\nu}^1(\mathbf{r}), \delta \tilde{n}_{\mathbf{q},\nu}^2(\mathbf{r}), \delta \tilde{n}_{\mathbf{q},\nu}^3(\mathbf{r}))$:

$$\begin{aligned} \left\{ \delta \tilde{n}_{\mathbf{q},\nu}^{\sigma,\sigma'}(\mathbf{r}) \right\} &\rightarrow \delta \tilde{n}_{\mathbf{q},\nu}(\mathbf{r}) = \delta \tilde{n}_{\mathbf{q},\nu}^0(\mathbf{r}) \sigma_0 + \delta \tilde{n}_{\mathbf{q},\nu}^1(\mathbf{r}) \sigma_1 \\ &\quad + \delta \tilde{n}_{\mathbf{q},\nu}^2(\mathbf{r}) \sigma_2 + \delta \tilde{n}_{\mathbf{q},\nu}^3(\mathbf{r}) \sigma_3 \quad (2) \\ &= \delta \tilde{n}_{\mathbf{q},\nu}^0(\mathbf{r}) \sigma_0 + \delta \tilde{\mathbf{M}}_{\mathbf{q},\nu}(\mathbf{r}) \boldsymbol{\sigma}. \end{aligned}$$

In real space, the time-dependent charge-spin field is given by the real part of the above frozen complex amplitudes when accounting for the classical motion of atoms. For a single phonon mode (\mathbf{q}, ν) of energy $\omega_{\mathbf{q},\nu}$ we have

$$\begin{aligned} \delta \mathbf{n}_{\mathbf{q},\nu}(\mathbf{r}, t) &= \text{Re} \left[\left(\delta \tilde{n}_{\mathbf{q},\nu}^0(\mathbf{r}) \sigma_0 + \delta \tilde{\mathbf{M}}_{\mathbf{q},\nu}(\mathbf{r}) \boldsymbol{\sigma} \right) e^{i(\mathbf{q}\mathbf{r} - \omega_{\mathbf{q},\nu} t)} \right] \\ &= \delta n_{\mathbf{q},\nu}^0(\mathbf{r}, t) \sigma_0 + \delta \mathbf{M}_{\mathbf{q},\nu}(\mathbf{r}, t) \boldsymbol{\sigma}, \end{aligned}$$

concluding that the appearance of an induced spin-density (or magnetization indistinctly) ($\delta \tilde{\mathbf{M}}_{\mathbf{q},\nu}(\mathbf{r})$) is completely general for crystals with spin-orbit coupling since the only requirement is a non-trivial pattern of the spin-polarization within the BZ associated to the absence of inversion symmetry⁵. The phonon modes break the symmetry of the BZ in a way that the electron spin-polarization is modulated within the BZ producing a net spin accumulation. The similarity of the phonon magnetism and the electron spin-polarization with time-reversal symmetry (no net magnetism) is strengthened by the fact that time-reversed phonon momenta give strictly opposite magnetization exactly in the same way as for electrons $\delta \mathbf{M}_{\mathbf{q},\nu}(\mathbf{r}, t) = -\delta \mathbf{M}_{-\mathbf{q},\nu}(\mathbf{r}, t)$.

The above description of the spin-charge field induced by phonons is completely classical and focussed on a single phonon with a fixed momentum. Therefore, physically $\delta \mathbf{M}_{\mathbf{q},\nu}(\mathbf{r}, t)$ would be the time dependent magnetization associated with a single coherent phonon mode. In general the vector field defined by $\delta \mathbf{M}_{\mathbf{q},\nu}(\mathbf{r}, t)$ shows an interesting real-space and time dependent non-collinear pattern, which depends also on the particular atomic displacements (polarization vectors) associated with each phonon branch. Actually, it is the motion of the W atom, i.e., the atom for which the spin-orbit interaction is dominant, which determines the direction of the magnetization. For instance, for $\mathbf{q} = \mathbf{K}$ and for a mode in which Se atoms rotate clockwise with opposite phase in the plane of the surface (x - y plane) and W atoms vibrate in the perpendicular direction (z) to the surface, a net circularly polarized induced magnetization appears in the surface plane around the W atoms (Fig. 1a-c and Supplementary Movie 1). However, for the same phonon propagation vector $\mathbf{q} = \mathbf{K}$, if W atoms rotate clockwise around their equilibrium positions in the plane of the surface, the net magnetization shows along the perpendicular direction to the plane (Fig. 2a-c and Supplementary Movie 2). It is noteworthy that the magnitude of the induced magnetization is only an order of magnitude smaller than in the induced (scalar) charge (Fig. 2b). Since $\mathbf{q} = \mathbf{K}'$ is the time-reversed momentum of $\mathbf{q} = \mathbf{K}$, as mentioned earlier, the real-space magnetization should be opposite in sign. For the first example given above but in the case in which $\mathbf{q} = \mathbf{K}'$, Se atoms rotate anticlockwise in the plane and the W vibrates perpendicular to the plane (Supplementary Movie 3). Taking

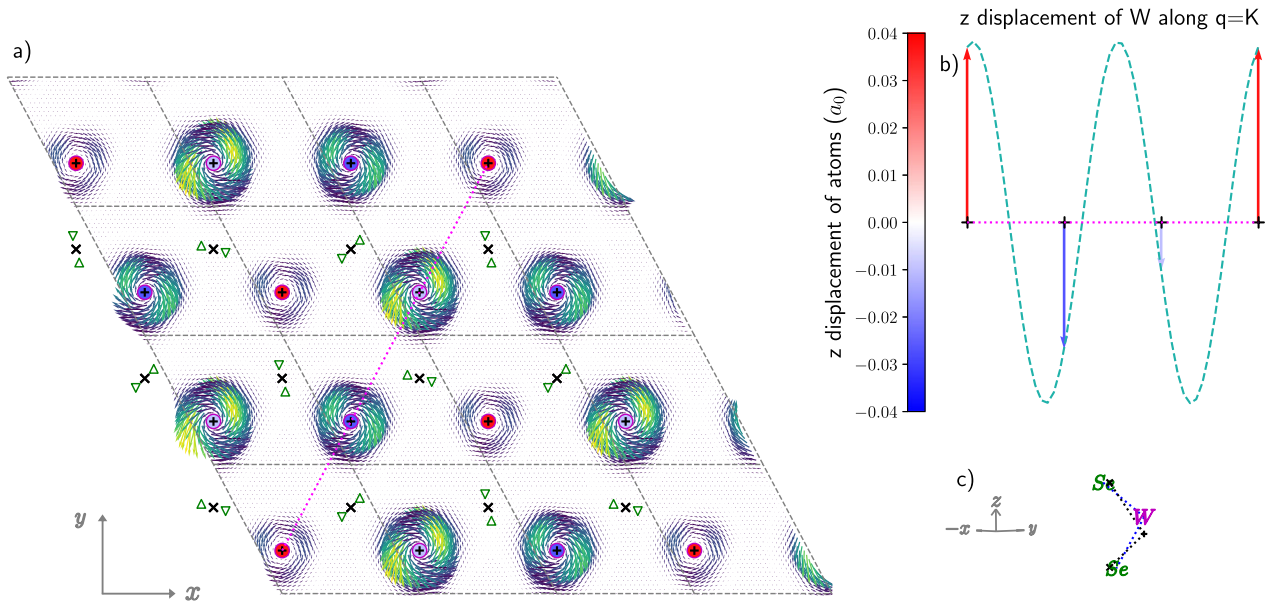


Fig. 1 Induced magnetization for the second lowest energy acoustic phonon mode at $\mathbf{q} = \mathbf{K}$ involving pure out of plane displacements of W atoms. **a** Real-space representation of the magnetization in the plane of the W atoms for 4×4 unit cells along the hexagonal axes of WSe_2 for the second lowest energy mode at $\mathbf{q} = \mathbf{K}$ (in orange in Fig. 3a, b). In this mode the W atoms (filled circles) displace along the perpendicular direction (see color-bar) and the Se atoms above (filled triangle up) and below (filled triangle down) the W plane rotate clockwise with opposite phase around their equilibrium positions (crosses) in their respective planes. The colored vector-field is proportional to the in-plane magnetization at each point in real space, with yellow/light (blue/dark) arrows representing the largest (smallest) values. These arrows as well as the displacements of the Se atoms have been scaled to make them visible. **b** The colored arrows give the z displacement of the W atoms along the $\mathbf{q} = \mathbf{K}$ direction (dotted magenta line in **a**) according to the color-bar. The dashed line describes the propagation of the vibration along several unit-cells in real space. Note that $\mathbf{K} = [1/3, 1/3, 0]$ in crystal axes, and hence the periodicity of the wave. **c** Side view of the WSe_2 formula-unit in the lower left corner unit-cell. The names of the atoms display their displacements from the equilibrium positions, denoted as in **a**. This figure is a snapshot of the time evolution of the induced magnetization for this mode (Supplementary Movie 1).

a snapshot in time for which the atomic positions coincide with those in Fig. 1 demonstrates that the magnetization shows exactly opposite chirality, and therefore the relation $\delta\mathbf{M}_{\mathbf{K},\nu}(\mathbf{r}, t) = -\delta\mathbf{M}_{-\mathbf{K},\nu}(\mathbf{r}, t)$ is fulfilled (Supplementary Fig. 1). Note that the propagation of the atomic displacements of W in the perpendicular direction to the plane along $\mathbf{q} = \mathbf{K}$ (Fig. 1b) is exactly the same as the one along $\mathbf{q} = \mathbf{K}'$ when it is looked from right to left (Supplementary Fig. 1b). A similar situation occurs for the second example, where the chirality of the atomic displacements reverses when changing $\mathbf{q} = \mathbf{K}$ to its time-reversed value⁶, which again gives a magnetization opposite in sign (Supplementary Movie 4) and which can be compared to that of Fig. 2 when the atomic positions are frozen to be the same (Supplementary Fig. 2). The non-collinear character in space and time of the magnetization is also observed when the atomic displacements are all linear. For instance, for an acoustic phonon with vector $\mathbf{q} = \mathbf{M}$ at which Se atoms move along \mathbf{q} and W atoms vibrate in the perpendicular direction to the plane (Supplementary Movie 5) a chiral magnetization pattern similar to that of Fig. 1 is found (Supplementary Fig. 3). Instead, when all atoms oscillate linearly in the plane at right angles to the propagation vector $\mathbf{q} = \mathbf{M}$ (Supplementary Movie 6), the magnetization appears in the perpendicular direction to the plane as in Fig. 2 (Supplementary Fig. 4). Note that for this acoustic mode when all atoms go through their equilibrium positions both, the induced charge and spin-density fields, disappear. All the magnetization patterns show a periodicity in real space according to the wave number \mathbf{q} of the propagation of the excited phonon, as they should. At this point it is worth mentioning that the magnetic polarization of the electron gas as described in this manuscript does not

have a relation with the angular momentum of the atoms as described by Zhang et al.⁷. In our theory linearly polarized phonons with null angular momentum give a finite and meaningful contribution to the magnetization. It is therefore clear that the physics described in⁷ is different and not connected to the spin response of the electron gas as described in the present work.

The size of the fluctuations of the real space unit-cell average of these oscillations gives an order of the magnitude of this effect, even though it does not capture all the detailed structure in real space. It is nevertheless physically meaningful, allowing to analyze the momentum and mode dependence at the same time, and making a connection with the possibility of experimental detection as it will be shown shortly. More specifically, for a given phonon \mathbf{q} the root-mean-square (RMS) of the time dependence of the periodic part of this quantity reflects the overall amplitude of the spin-density associated to a single phonon mode. For each cartesian component α we have (see Supplementary Note 2):

$$\overline{\delta M_{\mathbf{q},\nu}^\alpha} = \sqrt{\left\langle \left(\text{Re} \int_{\Omega} \frac{d^3\mathbf{r}}{\Omega} \left[\delta \tilde{M}_{\mathbf{q},\nu}^\alpha(\mathbf{r}) e^{-i\omega_{\mathbf{q},\nu} t} \right] \right)^2 \right\rangle_T}. \quad (3)$$

The above classical RMS amplitudes of the magnetization are directly connected to the charge-charge, spin-charge and spin-spin components of the dynamic structure factor, $S_{\alpha,\beta}(\omega, \mathbf{q} + \mathbf{G})$ (see Supplementary Note 2), which is accessed by inelastic neutron scattering, inelastic X-ray spectroscopy and spin-polarized electron energy loss spectroscopy⁸. As van Hove first pointed out⁹, the dynamic structure factor is the space and

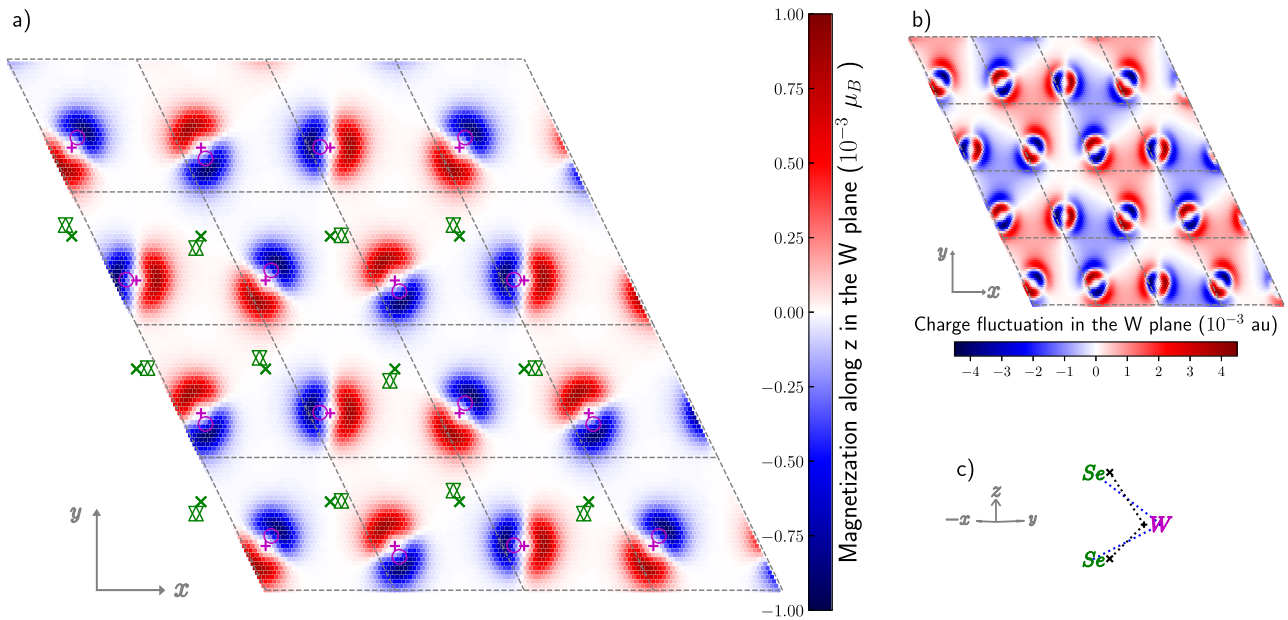


Fig. 2 Induced magnetization for the highest energy acoustic phonon mode at $\mathbf{q} = \mathbf{K}$ involving in-plane displacements of the W atoms. **a** Real-space representation of the perpendicular component of the magnetization in 4×4 unit cells along the hexagonal axes of WSe_2 for the highest energy acoustic mode for $\mathbf{q} = \mathbf{K}$ (represented in green in Fig. 3a, c. This mode is composed by clockwise rotations of the W atoms (circles) around their equilibrium positions and in-phase anticlockwise rotations of the Se atoms located above (triangle up) and below (triangle down) the W plane. The color code represents the magnetization in the perpendicular direction. The displacements of the atoms have been scaled to make them visible. **b** Induced electronic charge for the same atomic configuration as in panel **a**. Note that the induced magnetization is only an order of magnitude smaller than the induced (scalar) charge. **c** Side view of the WSe_2 formula-unit in the lower left corner unit-cell. The names of the atoms display their displacements from the equilibrium positions, denoted as in **a**. This figure is a snapshot of the time evolution of the induced magnetization for this mode (Supplementary Movie 2).

time Fourier transform of the density–density correlation function $\langle \delta \hat{\mathbf{n}}^\alpha(\mathbf{r}, t) \delta \hat{\mathbf{n}}^\beta(\mathbf{r}', 0) \rangle_T$. If we consider a 4-dimensional spin-charge quantized field

$$\delta \hat{\mathbf{n}}(\mathbf{r}, t) = \sum_{\mathbf{q}, \nu} \left(a_{\mathbf{q}, \nu}^\dagger e^{i\omega_{\mathbf{q}, \nu} t} \delta \tilde{\mathbf{n}}_{\mathbf{q}, \nu}^*(\mathbf{r}) + a_{\mathbf{q}, \nu} e^{-i\omega_{\mathbf{q}, \nu} t} \delta \tilde{\mathbf{n}}_{\mathbf{q}, \nu}(\mathbf{r}) \right), \quad (4)$$

where $a_{\mathbf{q}, \nu}$ and $a_{\mathbf{q}, \nu}^\dagger$ are creation and annihilation operators for a phonon mode (\mathbf{q}, ν) with energy $\omega_{\mathbf{q}, \nu}$ (Fig. 3a), then the dynamic structure factor can be written as

$$S_{\alpha, \beta}(\omega, \mathbf{q} + \mathbf{G}) = [1 + f_B(\omega_{\mathbf{q}, \nu})] \delta \tilde{n}_{\mathbf{q}, \nu}^\alpha(\mathbf{G}) \delta \tilde{n}_{\mathbf{q}, \nu}^{\beta*}(\mathbf{G}) \delta(\omega - \omega_{\mathbf{q}, \nu}) + f_B(\omega_{\mathbf{q}, \nu}) \delta \tilde{n}_{\mathbf{q}, \nu}^{\alpha*}(\mathbf{G}) \delta \tilde{n}_{\mathbf{q}, \nu}^\beta(\mathbf{G}) \delta(\omega + \omega_{\mathbf{q}, \nu}), \quad (5)$$

where $\delta \tilde{n}_{\mathbf{q}, \nu}^\alpha(\mathbf{G})$ indicates the Fourier transform or crystal field components of the real-space complex amplitudes $\delta \tilde{n}_{\mathbf{q}, \nu}^\alpha(\mathbf{r})$, $f_B(\omega_{\mathbf{q}, \nu})$ denotes phonon occupation numbers and where we ignore the Debye-Waller factor¹⁰. Taking the $\mathbf{G} = \mathbf{0}$ components (unit-cell average), it is easily seen that the classical RMS terms of Eq. (3) are proportional to the diagonal ($\alpha = \beta$) spectral contributions to Eq. (5) for individual phonons: $(\overline{\delta M_{\mathbf{q}, \nu}^\alpha})^2 \sim \delta \tilde{n}_{\mathbf{q}, \nu}^{\alpha*}(\mathbf{0}) \delta \tilde{n}_{\mathbf{q}, \nu}^\alpha(\mathbf{0})$. This helps to physically interpret the RMS of the induced magnetization defined as above because the terms parallel ($\sqrt{(\overline{\delta M_{\mathbf{q}, \nu}^x})^2 + (\overline{\delta M_{\mathbf{q}, \nu}^y})^2}$) and perpendicular ($\overline{\delta M_{\mathbf{q}, \nu}^z}$) to the WSe_2 layer shown in Fig. 3b, c, respectively, are basically the spin contributions to the structure factor connected to a given phonon mode (\mathbf{q}, ν) . Said in other words, Fig. 3b, c may be interpreted as the momentum/energy and phonon mode resolved contributions to the spin sector of the dynamic structure factor depicted along the high symmetry

lines of the surface Brillouin zone. The magnetic character associated inherently to phonons as proposed in this work is therefore accessible by means of any experimental setup probing the spin components of the dynamical structure factor in the energy ranges corresponding to phonons.

Discussion

We conclude that in any material with a non-trivial spin-pattern within the Brillouin zone, even if non-magnetic, phonon modes are connected inherently to a magnetic property analogous to the electron spin-polarization and it can be stated quite generally that phonon modes are accompanied by an induced spin-density (magnetization), which is rich in real space details. It is also shown that this magnetic modulation is only one order of magnitude weaker than the purely electrostatic (spin-diagonal) terms. All the above physics is illustrated convincingly for WSe_2 in a mode by mode analysis where the real space and time dependence of the induced magnetization is revealed for the most relevant modes. The implications are extensive and profound because phonons, which are now intrinsically attached to an effective magnetic moment, should be understood as an additional spin-flip mechanism even for materials without a net magnetic moment. This means that the whole electron–phonon physics is modulated in every system with spin–orbit coupling and, for instance, even electron backscattering events may be aided by the phonon magnetic moment. Experimental detection of magnetic oscillations for coherent phonons should be done by ultrafast probes and our calculated details of these fields may indicate a detection strategy. However, we also show that probing the spin components of the dynamical structure factor may be an alternative route to measure what we could name as the spin polarization of phonons.

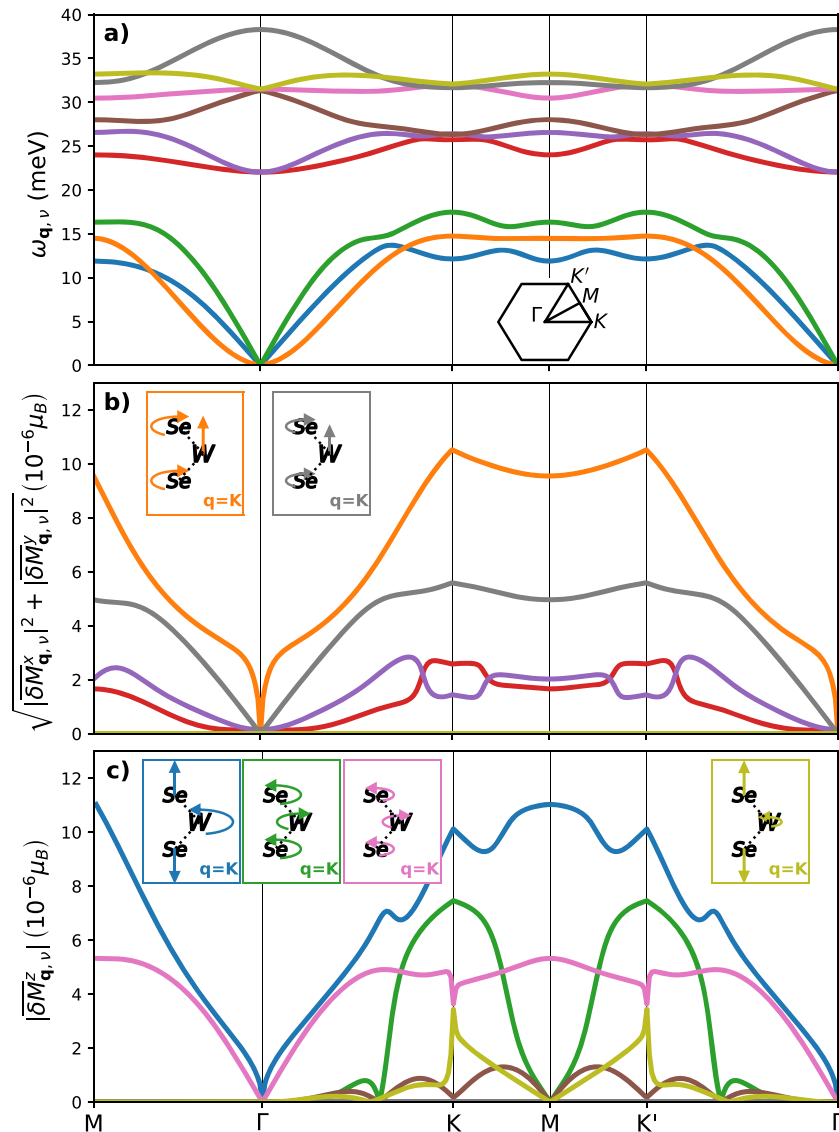


Fig. 3 Mode and momentum resolved magnetization induced by lattice vibrations in monolayer WSe₂. **a** Phonon energy spectrum along high symmetry lines in the surface Brillouin zone (inset). **b, c** Plane (*x, y*) and out-of-plane (*z*) components of the magnitude of the unit-cell average magnetization for each phonon mode in **a** and with the same colour convention. Insets show the corresponding polarization vectors for $q = K$, the length of the arrows being proportional to the magnitude of the atomic displacements. Vertical arrows represent linear displacements in the perpendicular direction to the plane, and semicircular arrows show circular displacements of the atoms around their equilibrium positions. The direction of the magnetization is determined by the motion of the W atom. When W vibrates in the perpendicular direction of the plane, it induces a net magnetization in the plane (**b**). However, when W atoms move on the plane (with circular polarization for $q = K$), the induced magnetization appears in the perpendicular direction (**c**). For the three middle modes in the phonon spectrum W atoms move significantly less, yielding a smaller magnetization. Panels **b, c** can be interpreted as a frequency and momentum resolved dynamic structure factor of phonons.

Data availability

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

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Author contributions

A.E. conceived the ideas. A.I. contributed to the early stages of the calculations. I.G.G. and A.E. carried out the calculations, discussed the results and contributed to the writing of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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