Angular Momentum of Phonons and the Einstein–de Haas Effect

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We study the angular momentum of phonons in a magnetic crystal. In the presence of a spin-phonon interaction, we obtain a nonzero angular momentum of phonons, which is an odd function of magnetization. At zero temperature, a phonon has a zero-point angular momentum in addition to a zero-point energy. With increasing temperature, the total phonon angular momentum diminishes and approaches zero in the classical limit. The nonzero phonon angular momentum can have a significant impact on the Einstein–de Haas effect. To obtain the change of angular momentum of electrons, the change of the phonon angular momentum needs to be subtracted from the opposite change of the lattice angular momentum. Furthermore, the finding of the phonon angular momentum gives a potential method to study the spin-phonon interaction. Possible experiments on phonon angular momentum are also discussed.

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The Einstein–de Haas effect [1,2], a phenomenon of mechanical rotation induced by a magnetization change, was originally designed to prove the existence of Ampere's molecular currents, but subsequent experiments [3] showed that the magnetic moment of an atom is dominated by spin while contribution from orbital motion to the magnetic moment is almost absent. The Einstein–de Haas experiment together with the Barnett experiment [4,5] (a change of magnetization resulting from a mechanical rotation) has provided an effective method of measuring the gyromagnetic ratio for various materials [6–8]. The accuracy of gyromagnetic ratio is crucial to determining of orbital and spin contribution in total magnetization [9–15].

Because of conservation of total angular momentum of the whole system in the Einstein–de Haas effect, the change of angular momentum of electrons (including both spin and orbital parts) is taken to be equal in magnitude but opposite in sign to the change of lattice angular momentum, which corresponds to mechanical rotation. However, the mechanical rotation only reflects angular momentum of the rigid-body lattice where atoms are assumed in the corresponding equilibrium positions, while phonons, which come from atomic vibrations around equilibrium positions, are assumed to have no macroscopic angular momentum. Recently, a remarkable phenomenon of the phonon Hall effect was observed in a paramagnetic insulator [16,17], which is indeed a surprise since phonons as neutral quasiparticles cannot directly couple to magnetic field via Lorentz force. The following theoretical studies [18,19] showed that through Raman spin-phonon interaction the magnetic field can have an effective force to distort phonon transport, and thus drive a circulating heat flow [20]. Therefore, a natural question arises: can such circulating phonons have nontrivial angular momentum and emergent macroscopic effects?

In this Letter, we study the angular momentum of phonons in a magnetic crystal in a microscopic picture. It is found that the Raman spin-phonon interaction induces a nonzero phonon angular momentum, which is an odd function of magnetization. In addition to a zero-point energy, the phonon has a zero-point angular momentum at zero temperature. Such zero-point phonon angular momentum is offset by that of excited phonon modes such that the total angular momentum of phonons vanishes in the classical limit. Phonon angular momentum cannot be ignored in total angular momentum especially in magnetic materials with large magnetization and spin-phonon interaction. Revisiting the Einstein–de Haas effect, we find that phonon angular momentum needs to be subtracted in calculating the angular momentum of electrons. With this correction, the spin and orbital angular momentum can be precisely determined. In addition to the Einstein–de Haas effect, nontrivial phonon angular momentum can be applied to the study of spin-phonon interaction, thermal Hall effect, and other topics related to phonons.

Angular momentum of phonons.—The lattice angular momentum related to mechanical rotation only reflects the rigid-body motion of the lattice. However, the angular momentum of phonons has never been considered. In a microscopic picture, we can define the angular momentum of phonons as

$$\boldsymbol{J}^{\mathrm{ph}} = \sum_{l\alpha} \boldsymbol{u}_{l\alpha} \times \dot{\boldsymbol{u}}_{l\alpha}. \tag{1}$$

Here $\boldsymbol{u}_{l\alpha}$ is a displacement vector of the αth atom in the lth unit cell, multiplied by square root of mass. Along z direction, $J_z^{\mathrm{ph}} = \sum_{l\alpha}(u_{l\alpha}^x \dot{u}_{l\alpha}^y - u_{l\alpha}^y \dot{u}_{l\alpha}^x)$. One can present the displacement in the second quantization form as $u_l = \sum_k \varepsilon_k e^{i(\mathbf{R}_l \cdot \boldsymbol{k} - \omega_k t)} \sqrt{\frac{\hbar}{2\omega_k N}} a_k + \mathrm{H.c.}$, with $k = (\boldsymbol{k}, \sigma)$

specifying a wave vector $\boldsymbol{k}$ and a branch σ, where ϵ_k is a displacement polarization vector. Then the phonon angular momentum can be written as [21]

$$J_z^{\mathrm{ph}} = \frac{\hbar}{2}\sum_{k,k'} \epsilon_k^\dagger M \epsilon_{k'} \left(\sqrt{\frac{\omega_k}{\omega_{k'}}} + \sqrt{\frac{\omega_{k'}}{\omega_k}}\right) a_k^\dagger a_{k'} \delta_{\boldsymbol{k},\boldsymbol{k}'} e^{i(\omega_k - \omega_{k'})t} + \frac{\hbar}{2}\sum_k \epsilon_k^\dagger M \epsilon_k. \quad (2)$$

Here $M = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix} \otimes I_{n\times n}$, and n is the number of atoms in one unit cell. In equilibrium, the angular momentum of phonons reduces to [21]

$$J_z^{\mathrm{ph}} = \sum_{\sigma,\boldsymbol{k}} l_{\boldsymbol{k},\sigma}^z \left[f(\omega_{\boldsymbol{k},\sigma}) + \frac{1}{2}\right], l_{\boldsymbol{k},\sigma}^z = (\epsilon_{\boldsymbol{k},\sigma}^\dagger M \epsilon_{\boldsymbol{k},\sigma})\hbar, \quad (3)$$

where $f(\omega_k) = 1/(e^{\hbar\omega_k/k_BT} - 1)$ is the Bose-Einstein distribution. In Eq. (3), we do a summation over all wave vector points and all phonon branches ($\omega \geq 0$). Here, $l_{\boldsymbol{k},\sigma}^z$ is the phonon angular momentum of branch σ at wave vector $\boldsymbol{k}$, which is real and proportional to $\hbar$. At zero temperature, the total phonon angular momentum is $J_z^{\mathrm{ph}}(T=0) = \sum_{\sigma,\boldsymbol{k}} \frac{1}{2} l_{\boldsymbol{k},\sigma}^z$, which means that each mode of ($\boldsymbol{k}$, σ) has a zero-point angular momentum $\frac{1}{2} l_{\boldsymbol{k},\sigma}^z = (\hbar/2)(\epsilon_{\boldsymbol{k},\sigma}^\dagger M \epsilon_{\boldsymbol{k},\sigma})$ in addition to a zero-point energy of $\hbar\omega_{\boldsymbol{k},\sigma}/2$.

For an ionic crystal lattice in a uniform external magnetic field, the Hamiltonian reads in a compact form [18,19,22,23],

$$H = \tfrac{1}{2}(p - \tilde{A}u)^T(p - \tilde{A}u) + \tfrac{1}{2}u^T K u, \quad (4)$$

where u is a column vector of displacements from lattice equilibrium positions, multiplied by square root of mass; p is a conjugate momentum vector, and K is a force constant matrix. The cross term $u^T\tilde{A}p$ can be interpreted as a Raman spin-phonon interaction [24,25]. The superscript T stands for the matrix transpose. $\tilde{A}$, an antisymmetric real matrix [26], has a dimension of $Nd \times Nd$ where N is the number of total sites and d is the dimension of lattice vibrations; in a proper approximation it can be block diagonal with elements $\Lambda_\alpha = \begin{pmatrix} 0 & \lambda_\alpha \\ -\lambda_\alpha & 0 \end{pmatrix}$ with respect to the αth ionic site, where we only consider two-dimensional (x and y directions) motion of the lattice ($d = 2$). Here λ_α has a dimension of frequency, and is proportional to the spin-phonon interaction and magnetization, which is assumed to be proportional to magnetic field for a paramagnetic material. The magnetic field is applied along z direction. The polarization vector ϵ satisfies $[(-i\omega + A)^2 + D]\epsilon = 0$, where $D(\mathbf{k}) = -A^2 + \sum_{l'} K_{ll'} e^{i(\mathbf{R}_{l'} - \mathbf{R}_l)\cdot\mathbf{k}}$ is the dynamic matrix and A is block diagonal with the element of Λ_α, and has a dimension of $2n \times 2n$ where n is the number of sites per unit cell.

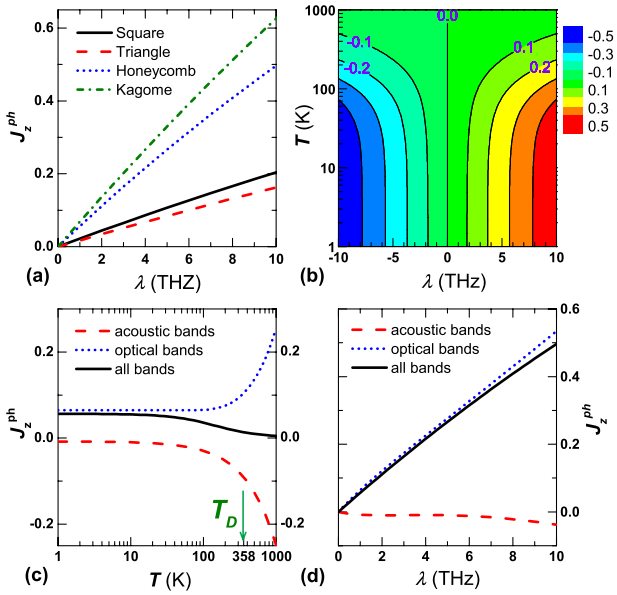

FIG. 1 (color online). (a) The phonon angular momentum J_z^{ph} of one unit cell as a function of λ at temperature $T = 0$ K for different lattice symmetries. (b) The contour plot of the phonon angular momentum J_z^{ph} of one unit cell as a function of λ and temperature T. (c) The phonon angular momentum J_z^{ph} of one unit cell from different phonon bands as a function of temperature T at $\lambda = 1$ THz, where the arrow denotes the Debye temperature of the model ($T_D = 358$ K). (d) The phonon angular momentum J_z^{ph} of one unit cell from different phonon bands as a function of λ at $T = 0$ K. The phonon angular momenta in (b)–(d) are calculated for a honeycomb lattice. All the phonon angular momenta are in the unit of $\hbar$.

In absence of spin-phonon interaction, the system reduces to a trivial phonon system $H = \frac{1}{2}p^T p + \frac{1}{2}u^T K u$. Solving the simple eigenvalue problem as $D(\boldsymbol{k})\epsilon_{\boldsymbol{k},\sigma} = \omega_{\boldsymbol{k},\sigma}^2 \epsilon_{\boldsymbol{k},\sigma}$ with $D^T(\mathbf{k}) = D^*(\mathbf{k}) = D(-\mathbf{k})$, one can have $\omega_{-\boldsymbol{k},\sigma} = \omega_{\boldsymbol{k},\sigma}, \epsilon_{-\boldsymbol{k},\sigma} = \epsilon_{\boldsymbol{k},\sigma}^*$, then we obtain $l_{-\boldsymbol{k},\sigma}^z = -l_{\boldsymbol{k},\sigma}^z$ and $J_z^{\mathrm{ph}} = 0$ [21]. Thus for a phonon system without a spin-phonon interaction, the total angular momentum of phonons is zero.

For a phonon system with a spin-phonon interaction, $\epsilon_{-\boldsymbol{k},\sigma} = \epsilon_{\boldsymbol{k},-\sigma}^* \neq \epsilon_{\boldsymbol{k},\sigma}^*$, and then $l_{-\boldsymbol{k},\sigma}^z \neq -l_{\boldsymbol{k},\sigma}^z$; thus, one can get a nonzero phonon angular momentum, which is shown in Fig. 1. We calculate phonon angular momentum for lattices with the following parameters: the longitudinal spring constant is $K_L = 0.144$ eV/(uÅ^2) and the transverse one is $K_T = K_L/4$; the unit cell lattice vectors are $(a,0)$, $(0,a)$ for a square lattice and $(a,0)$,$(a/2,a3/2)$ for other lattices with $a = 1$ Å. We take $\lambda_\alpha = \lambda$ for the model calculation [27]. Figure 1(a) shows that honeycomb and kagome lattices have larger phonon angular momenta than those of triangle and square lattices, which means that lattices with more sites per unit cell can have a larger phonon angular momentum. We can understand this trend

by observing that optical bands are more important in contributing to the phonon angular momentum than the acoustic ones. In Figs. 1(c) and 1(d) we plot the phonon angular momentum contributing from different bands in a honeycomb lattice. It is shown that the phonon angular momentum from acoustic bands almost vanishes at low temperatures [see Fig. 1(c)] and if λ is not large [see Fig. 1(d)]; thus, the optical bands dominate the contribution to the total phonon angular momentum. With more sites per unit cell more optical bands are present; thus, the phonon angular momentum will be larger.

By using the relations $\epsilon^*_{-\boldsymbol{k},\sigma}(-A)=\epsilon_{\boldsymbol{k},\sigma}(A)$, $\omega_{-\boldsymbol{k},\sigma}(-A)=\omega_{\boldsymbol{k},\sigma}(A)$, $M^T=-M$, we can obtain $J_z^{\rm ph}(-\lambda)=-J_z^{\rm ph}(\lambda)$. Since λ is proportional to magnetization, the total angular momentum of phonon will change sign when magnetization changes sign. As shown in Figs. 1(a), 1(b), and 1(d), the total angular momentum of phonons per unit cell increases as λ increases, but the increase rate will decrease.

Angular momentum in the classical limit.—At the high temperature limit, from Eq. (3) we have [21]

$$J_z^{\rm ph}(T\to\infty)=\sum_{\sigma>0,\boldsymbol{k}}\left[\left(\frac{k_BT}{\hbar\omega_{\boldsymbol{k},\sigma}}+\frac{\hbar\omega_{\boldsymbol{k},\sigma}}{12k_BT}\right)l^z_{\boldsymbol{k},\sigma}\right]. \quad (5)$$

It seems that the phonon angular momentum would be linear with temperature at the high temperature limit. However, the first term vanishes due to the fact of $\sum_{\sigma>0,\boldsymbol{k}}(\epsilon^\dagger_{\boldsymbol{k},\sigma}M\epsilon_{\boldsymbol{k},\sigma}/\omega_{\boldsymbol{k},\sigma})=0$ [21]. Therefore, at a high temperature the total phonon angular momentum is proportional to $1/T$ and tends to zero as

$$J_z^{\rm ph}(T\to\infty)=\sum_{\sigma>0,\boldsymbol{k}}\frac{\hbar\omega_{\boldsymbol{k},\sigma}}{12k_BT}l^z_{\boldsymbol{k},\sigma}\to 0. \quad (6)$$

The phonon angular momentum per unit cell changing with temperature is shown in Figs. 1(b) and 1(c). Whatever a magnetic field is applied, the phonon angular momentum per unit cell decreases with increasing temperature and tends to zero at the high temperature limit ($T\gg T_D$). With increasing temperature more modes are exited, the angular momentum of which has the direction opposite to that of the zero-point angular momentum; at the high temperature limit, the phonon angular momentum of all the excited modes exactly cancels out the zero-point angular momentum [$\sum l^z_{\boldsymbol{k},\sigma}f(\omega_{\boldsymbol{k},\sigma},T\to\infty)=-\sum\frac{1}{2}l^z_{\boldsymbol{k},\sigma}$]. We can understand the absent phonon angular momentum in the classical limit as follows. At high temperatures, classical statistical mechanics is applicable to calculate the phonon angular momentum. Summation over quantum states becomes a phase-space integral with respect to p and u. One can do a change of variable to make the kinetic energy in the Hamiltonian equation (4) into a usual form $p^2/2$, thus removing the effect of $\tilde{A}u$; for such a pure harmonic system, the angular momentum of phonons is zero as discussed above. Furthermore, the Bohr–van Leeuwen theorem states that in classical mechanics the thermal average of the magnetization is always zero [28], which also makes the angular momentum of phonons vanish at the classical limit. Therefore, the phonon angular momentum is meaningful only in low-temperature quantum systems.

Revisit the Einstein–de Haas effect—The Einstein–de Haas effect [1] showed a mechanical rotation of a freely suspended body caused by the change in its magnetization. In their experiment [1], Einstein and de Haas employed a resonance method in which the magnetic field was periodic and tuned to be the natural frequency of the rod and its suspension, which provided measurements for the ratio between the change in magnetization and the one in the total angular momentum. Traditionally the total angular momentum is assumed as $\boldsymbol{J}^{\rm tot}=\boldsymbol{J}^{\rm lat}+\boldsymbol{J}^{\rm spin}+\boldsymbol{J}^{\rm orb}$; thus, due to conservation of the angular momentum, one obtains $\Delta\boldsymbol{J}^{\rm lat}=-(\Delta\boldsymbol{J}^{\rm spin}+\Delta\boldsymbol{J}^{\rm orb})$, which is determined by the mechanical rotation of the sample [7]. However, from a microscopic point of view, the angular momentum of all atoms in the sample can be written as

$$\boldsymbol{J}^{\rm atom}=\sum_{l\alpha}(\boldsymbol{R}_{l\alpha}+\boldsymbol{u}_{l\alpha})\times(\dot{\boldsymbol{R}}_{l\alpha}+\dot{\boldsymbol{u}}_{l\alpha}), \quad (7)$$

where $\boldsymbol{R}_{l\alpha}$ is the equilibrium position of the αth atom in the lth unit cell, multiplied by square root of its mass. The angular momentum of the lattice is

$$\boldsymbol{J}^{\rm lat}=\sum_{l\alpha}\boldsymbol{R}_{l\alpha}\times\dot{\boldsymbol{R}}_{l\alpha}, \quad (8)$$

which really reflects the mechanical rotation of rigid-body motion of the sample. In equilibrium, the cross terms related with $\boldsymbol{u}$ or $\dot{\boldsymbol{u}}$ are zero, then $\boldsymbol{J}^{\rm atom}=\boldsymbol{J}^{\rm lat}+\boldsymbol{J}^{\rm ph}$. Thus the total angular momentum should be

$$\boldsymbol{J}^{\rm tot}=\boldsymbol{J}^{\rm lat}+\boldsymbol{J}^{\rm ph}+\boldsymbol{J}^{\rm spin}+\boldsymbol{J}^{\rm orb}. \quad (9)$$

The global conservation of the angular momentum does not explain how the angular momentum is actually transferred from individual electrons or atoms to the whole rigid body; the Raman type spin-phonon interaction can be ubiquitous and plays an essential role. According to the discussion in the above section, we know that in the presence of the spin-phonon interaction, the phonon band structure is nontrivial and gives the nonzero angular momentum $\boldsymbol{J}^{\rm ph}$. Based on conservation of the total angular momentum, we obtain

$$\Delta\boldsymbol{J}^{\rm spin}+\Delta\boldsymbol{J}^{\rm orb}=-\Delta\boldsymbol{J}^{\rm lat}-\Delta\boldsymbol{J}^{\rm ph}. \quad (10)$$

Therefore, to obtain the change of the angular momentum of electrons, one needs to subtract the contribution of the phonon from the opposite change of the lattice angular momentum. On the other hand, one can measure the total magnetization change as

$$\Delta M = \Delta M^{\text{spin}} + \Delta M^{\text{orb}}. \quad (11)$$

Combining Eq. (10), Eq. (11) together with the facts of $\Delta M^{\text{orb}} = (e/2m)\Delta J^{\text{orb}}$ and $\Delta M^{\text{spin}} = (e/m)\Delta J^{\text{spin}}$, one can easily determine ΔM^{spin} and ΔM^{orb}.

The phonon can make a significant contribution to total angular momentum, while the magnitude of the phonon angular momentum depends on the value of λ. The parameter λ can be obtained from phonon dispersion relation since our calculation shows that in the presence of spin-phonon interaction degenerate phonon modes split at Γ point with a gap of 2λ. By means of Raman scattering experiments, literatures [29,30] show that the phonon splitting ranges up to about 26 cm^{-1} in paramagnetic CeF_3 at $T = 1.9$ K and $B = 6$ T; thus, λ can be about 0.39 THz and the phonon angular momentum per unit cell is about 0.02 $\hbar$. One also can estimate the parameter λ from the phonon Hall effect. For a paramagnetic terbium gallium garnet $Tb_3Ga_5O_{12}$, the parameter λ is estimated as $\lambda = 0.1$ $\text{cm}^{-1} \simeq 3$ GHz at $B = 1$ T and $T = 5.45$ K [18]; thus, in such material the phonon angular momentum per unit cell is about $1.6 \times 10^{-4}\hbar$, which is relatively small. However, one can observe a much larger phonon angular momentum when magnetization is saturated in this paramagnetic material since the parameter λ is proportional to magnetization. In the phonon Hall effect experiment [16,17], the paramagnetic insulator was chosen to manifest the phonon contribution in the thermal transport where the contribution from electron and magnon can be neglected. However, the spin-phonon interaction is widely present in various magnetic materials [31–34]. Ferromagnetic materials have very large magnetization; thus, one can expect a large phonon angular momentum. One also can observe evident phonon angular momentum in materials with strong spin-phonon interaction by using Raman spectroscopy, such as La_2NiMnO_6 [35], Sr_2CoO_4 [36] and cupric oxide [37].

Thus for materials with strong spin-phonon interaction together with large magnetization, the zero-point angular momentum of phonons can be significant. According to previous studies, in some ferromagnetic materials the calculated orbital magnetic moment is only a few percent of the total magnetic moment, that is, the orbital angular momentum is also around a few percent of $\hbar$ [8]; thus, the phonon angular momentum cannot be ignored. With improvement of experimental technique in past decades, the accuracy of the measurement has been much enhanced; thus, the phonon angular momentum should be measurable.

Possible experiment to separate the phonon angular momentum.—One can do experiments on a ferromagnetic insulator with saturation magnetization, where electron transport can be ignored. Due to the properties of the phonon angular momentum—it decreases with increasing temperature and vanishes in the classical limit—one can measure the change of the lattice angular momentum at low and high temperatures to separate the phonon angular momentum from the others. Here the temperature scale should be the Debye temperature which divides the quantum and classical regions. On the other hand, in order to avoid the involvement of magnons, we need to do experiments at temperatures that are low compared to the Curie temperature. This demands that the Curie temperature be much higher than the Debye temperature. Thus, the angular momentum of magnons almost keeps constant, while that of phonons changes dramatically with changing temperature. Fortunately, this can be satisfied by many ferromagnetic materials where their Curie temperature is around 1000 K while their Debye temperatures is less than 500 K [38].

In addition to its application to the measurement of gyromagnetic ratio, the nontrivial phonon angular momentum provides us a possible efficient route to study spin-phonon interaction in magnetic materials. On the other hand, how to separate the contribution from phonons and magnons to the thermal Hall effect in ferromagnetic materials is an open problem. The phonon angular momentum, however, can give a way to obtain the phonon contribution.

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