Topological phonons in an inhomogeneously strained silicon-3: Flexoelectronic
doping mediated topological electronic magnetism of phonons and Mott
transition

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Abstract

In metal/highly doped Si bilayer structure, strain gradient lead to charge carrier transfer from metal layer to the Si layer from interfacial flexoelectronic effect and possibly due to gradient in band structure and mobility, which is called as flexoelectronic doping. In addition, the strain gradient also give rise to topological phonons in Si due to inhomogeneity. The superposition of the topological phonons and flexoelectronic polarization is expected to give rise to topological electronic magnetism of phonons. In this study, we use magneto transport measurements to present experimental evidence of topological electronic magnetism. We use longitudinal resistance measurement as a function of temperature and current bias to uncover the large flexoelectronic doping. The charge carrier concentration in Si increases from $4.14 \times 10^{19}$ cm$^{-3}$ to $1.58 \times 10^{21}$ cm$^{-3}$ creating a charged Si, which led to large Coulomb repulsion and Mott metal insulator transition in Si. The topological electronic magnetism is uncovered using giant magnetoresistance behavior in the angle dependent magnetoresistance response. The superposition of flexoelectronic effect and topological phonons give rise to oscillatory Hall effect, which is attributed to the superposition of the spin-splitting from topological electronic magnetism and Zeeman splitting from external magnetic field. The flexoelectronic doping and topological electronic magnetism of phonons can lead a new class of magnetoelectronic multiferroic materials for energy efficient device applications. The flexoelectronic doping can also lead a potential pathway to high temperature superconductivity.
In a recent discovery, a large phonon magnetic moment (1.2 $\mu_B$/atom) was reported in Si thin film under an applied strain gradient. The large magnetic moment was attributed to the dynamical multiferroicity[1], which can be described as:

$$M_t \propto P_{FE} \times \partial_t P$$  \hspace{1cm} (1)$$

where $M_t$, $P$ and $P_{FE}$ are temporal magnetic moment and polarization of optical phonons and is the flexoelectronic effect. However, the measured phonon magnetic moment in was four orders of magnitude larger than that predicted according to circular motion of ions. Previously, Cheng et al.[2] also reported a large phonon magnetic moment in a Dirac semimetal Cd$_3$As$_2$. Ren et al.[3] proposed an alternate topological origin of the phonon magnetic moment to explain this behavior. The electronic topological magnetism arises due to phonon modified electronic energy in addition to k-resolved Born effective charge. The electronic topological magnetism is described as:

$$M_{qB} = \frac{e}{2m_i} L \int \frac{dk}{(2\pi)^2} \Omega_{ka} q_{ij} u_i u_j$$  \hspace{1cm} (2)$$

where $M_i$ is the mass of the representative ion with average angular momentum $L_i = \frac{m_i}{T} \int_0^T (u \dot{u})_z dt$ over the phonon period $T$, $\Omega$ is Berry curvature, $k$ is the momentum and $u_i$ is a displacement vector multiplied by the square root of the mass, and this vector is related to the displacement polarization vector[4]. It was noted that even electronic topological magnetism predicted an order of magnitude smaller value for the phonon magnetic moment. For the topological phonons in an inhomogeneously strained Si, the temporal magnetic moment (or spin angular momentum) of topological phonons can be described as:
\[ M_t \propto P_{FE} \times f(A, p) \] (3)

where \( A \) and \( p \) are Berry gauge potential and momentum, respectively. This relationship can be considered an alternate form of the topological magnetism where phonon Berry curvature replaces electronic Berry curvature. However, the large magnitude of phonon magnetic moment can only arise if the flexoelectronic effect is large.

In the Si thin films, applied strain gradient changes the position of valence band maxima as well as conduction band minima in addition to band gap. In case of lightly doped Si, the slope of the band structure will lead to charge separation and flexoelectronic effect as shown in Figure 1 (a). Whereas, in case of highly doped Si, the charge carrier will screen the flexoelectronic effect as shown in Figure 1 (a) and no net polarization will exist in the thin film. However, in the metal-Si (highly doped) bilayer structure, interfacial flexoelectronic effect[5] will also appear as shown in Figure 1 (b). The interfacial flexoelectronic effect will lead to charge carrier injection from metal layer to the Si layer as shown in Figure 1 (c). For example, the average charge carrier density in Si layer may increase from say \( 5 \times 10^{19} \) cm\(^{-3} \) to \( 7 \times 10^{19} \) cm\(^{-3} \), which will give rise to charge polarized Si.

It is noted that Wang et al. [5] did not report interfacial flexoelectronic effect in their highly doped Si samples because they measured only the I-V characteristics, which did not change.

Due to the charge carrier injection, the Si layer in the bilayer structure is no longer charge neutral. In addition, the charge carrier density will be asymmetric, which will induce a potential in the bulk of the Si layer equal in magnitude and opposite in sign to the
interfacial flexoelectronic effect as shown in Figure 1 (c) and flexoelectronic effect can be described as:

\[ P_{FE} \propto \frac{\partial n}{\partial z} \]  \hspace{1cm} (4)

where \( n(x,y,z) \) is spatially inhomogeneous charge carrier density. The flexoelectronic polarization will not be aligned with the strain gradient direction instead it will be aligned along the crystallographic direction having the largest linear density in the cross-sectional plane. The charge carrier injection from metal will also change the Fermi energy and electronic behavior of the metal layer as well. Our hypothesis is supported by a recent report where transition from electron to hole transport is observed in Pt/p-Si bilayer structure due to flexoelectronic effect. Wang et al. showed that the flexoelectronic effect in Si\(^5\) is almost two orders of magnitude larger than theoretical values for conventional flexoelectric effect in Si. The flexoelectronic effect in centrosymmetric materials can also be comparable to the magnitude of polarization in ferroelectric materials.\(^{6-8}\)

Now, the superposition of topological phonons and asymmetric charge carrier density due to flexoelectronic effect is expected to give rise to large magnetic moment as shown in Figure 1 (d) since the flexoelectronic effect arises due to the gradient of free charge carriers instead of ions. We can call it as topological electronic dynamical multiferroicity or topological electronic magnetism of phonons, which is described as:

\[ M_t \propto \frac{\partial n}{\partial z} \times f(A,p) \]  \hspace{1cm} (5)

Traditionally, the optical phonons having circular polarization are expected to have spin angular momentum. However, both transverse and longitudinal acoustic phonons may
also acquire spin angular momentum due to either superposition with optical phonons or from topological Berry gauge potential. The topological electronic dynamical multiferroicity can potentially give rise to conducting multiferroic materials. To support our hypothesis, we needed experimental evidence of flexoelectronic doping, topological magnetism and the effect due to superposition of flexoelectronic doping and topological phonons.

Figure 1. Figure 1. (a) schematic showing approximate band diagram of p-doped Si thin film due to flexoelectronic charge separation. (b) Schematic showing the band structure of metal/p-Si bilayer structure with interfacial flexoelectronic voltage, (c) schematic showing the charge carrier injection and flexoelectronic doping in the Py/Si bilayer thin film. (d) schematic showing the superposition of the charge carrier gradient due to flexoelectronic doping and topological phonons giving rise to topological electronic...
magnetism of phonons. (e) A scanning electron micrograph showing the experimental scheme and freestanding sample structure.

In our previous studies, we have used a commercially available SOI wafer with 2 µm device layer (0.001-0.005 Ω cm) and only method to modify the strain gradient was using increased Joule heating from larger current. In this study, we used chemical etching of the device layer of the SOI wafer to achieve the thickness closer to ~400 nm by successively oxidizing and, then, etching the thermal oxide using hydrofluoric (HF) acid[10]. The deposition of MgO (1.8 nm) and Ni_{80}Fe_{20} (Py) (25 nm) led to larger strain gradient in the 400 nm p-Si layer as compared to 2 µm layer. In addition, the strain gradient in the sample structure was further changed using an applied current bias, where self-heating changed the thermal mismatch stresses leading to enhanced bending inside a Quantum Design PPMS chamber. In the first experimental study, we measured the longitudinal resistance as a function of temperature from 350 K to 5 K at different applied current bias. The measured response is shown in Figure 2 (a). The measured responses changed as the applied current was increased from 10 µA to 2 mA as shown in Figure 2 (a). These responses were significantly different from the Py and p-Si responses. Most importantly, we observed a metal to insulator transition (MIT) below 100 K in all the measurements as shown in Figure 2 (b). The MIT response was expected to arise in Si layer only and, as a consequence, the residual resistance at 5 K was from the Py layer only as shown in Figure 2 (b). Additionally, the residual resistance after MIT was larger at higher currents whereas the resistance at 350 K was smaller at higher current as shown in Figure 2 (b). We attributed this behavior to the flexoelectronic doping (charge carrier injection) of the Si layer from Py layer due to interfacial flexoelectronic effect.
Figure 2. (a) the resistance response in Py (25 nm)/MgO/p-Si (400 nm) composite sample as a function of temperature and current bias from 10 µA to 2 mA, (b) the resistance response between 50 K and 5 K showing the Mott metal insulator transition in the Si layer and residual resistance of Py layer, (c) the estimated resistivity of the Si layer in the composite sample after subtracting Py response, and (d) a schematic showing the flexoelectronic doping of the Si layer due to charge carrier transfer from Py layer to Si layer.
The Py resistance response as a function of temperature from 300 K to 5 K was reported previously[10]. We used that data to simulate the resistivity using a cubic equation. The resistance response as a function of temperature from the Py layer would not change since it was a metal. From the residual resistance, we attributed the change in Py resistance as a function of current to flexoelectronic charge transfer or reduction in charge carrier concentration. For example, the resistance of the Py layer was expected to be ~ 185.17 Ω at 5 K as shown in Supplementary Figure S1 whereas the residual resistance was ~ 245.8 Ω at 10 µA. Hence, we multiplied the Py resistivity with a factor of 1.4 to simulate the resistance behavior at 10 µA of current. This process was repeated for each current. We, then, subtracted the Py resistivity response from the sample resistance to extract the resistivity behavior of Si layer, which is shown in Figure 2 (c). The estimated resistivity of the p-Si layer was an order of magnitude larger than the wafer resistivity, which was a clear indicator of flexoelectronic doping.

We assume that the mobility of charge carrier in Py layer will not change and any change in the resistivity of the Py layer will be due to reduction in the charge carrier concentration as stated earlier. We, then, inferred that the residual resistance of Py layer after MIT was due to reduction in charge carrier concentration. Using \( n_{Py} = 6.85 \times 10^{22} \text{ cm}^{-3} \), we estimated the change in charge carrier concentration for each current dependent measurement presented in Figure 2 (a). For example- the charge carrier concentration was expected to be \( 1.96 \times 10^{22} \text{ cm}^{-3} \) at 10 µA of current. The difference in charge carrier concentration was then transferred to the Si layer due to interfacial flexoelectronic effect. The new charge carrier concentration of Si at 10 µA of current was estimated to be \( 1.26 \times 10^{21} \text{ cm}^{-3} \) as compared to \( \sim 4.14 \times 10^{19} \text{ cm}^{-3} \) measured from Si (2 µm) sample as
shown in Figure 2 (d). The flexoelectronic doping reduced the overall resistance because the mobility of electrons in Py (~1.7 cm²/(V.s)) was an order of magnitude smaller than p-Si (~35 cm²/(V.s)). The measured mobility value was much smaller than previously reported bulk values because Si was degenerately doped. It is noted that strain gradient will also cause gradient of mobilities through the thickness of the sample. However, its effect on the charge transport and flexoelectronic doping was not well understood. The change in charge carrier concentration was calculated for each current as shown in Supplementary Table S1. We also measured the resistance behavior at 1 mA and out of plane magnetic field of 1 T as shown in Figure 2 (a-c). Based on the residual resistance of Py layer, the flexoelectronic doping was same in both cases but the temperature dependent response was significantly different. The external magnetic field led to the spin polarization of charge carrier in Py and in turn the injected charge carriers were also spin polarized, which was expected to enhance the topological magnetism of phonon. As a consequence, the exchange interactions led to changed resistance response observed in our measurement. Based on the number of charge carrier injection, we also estimated the maximum charge accumulation in Si layer to be 1250 C/m² at 10 µA of current, which was four orders of magnitude larger than the value reported recently by Wang et al. This value was also three orders of magnitude larger than ferroelectric polarization. The dynamical multiferroicity predicted the phonon temporal magnetic moment to be 10⁻⁴ µB whereas phonon magnetic moment of 1.2 µB/atom was reported in doped Si. While the estimated value of the charge accumulation might seem exceptionally large but without such a charge accumulation the experimentally observed large magnetic moment will also not arise. Our calculations replicated the resistance response and presented the first
experimental evidence of large flexoelectronic charge transfer responsible for the
topological magnetism of phonons.

A careful qualitative analysis suggested transitions in the Si layer as observed in
the resistance response as a function of temperature shown in Figure 2 (a,c). A
predominant MIT behavior was observed in all the measurements below 100 K. Another
MIT like behavior was observed at 200 K in the measurement at 10 µA current bias. In
Si, MIT occurs when the charge carrier concentration(doping) is below \(\sim 10^{18} \text{ cm}^{-3}\).
However, in the present study, the charge carrier concentration was an order of
magnitude larger than that even if the flexoelectronic doping was ignored. As described
earlier, the interfacial flexoelectronic effect led to charge carrier injection in the Si layer,
which was the underlying cause of large flexoelectronic polarization. At higher
temperatures, it gave rise to topological magnetism and exchange interactions, which
was potentially the underlying cause of MIT observed at 10 µA current as shown in Figure
2 (a). Since Si layer was not charge neutral due to excess charge carriers from
flexoelectronic doping; the large flexoelectronic polarization led to strong Coulomb
repulsion and opening a gap, which led to Mott transition at temperature below 100 K as
shown in Figure 2 (a-c). The resulting Mott transition can also be called as magneto-
electronic multiferroic insulator.
Figure 3. The Hall resistance measurement for an applied magnetic field of 8 T to -8T at 200 K showing (a) negative Hall resistance in Py (25 nm)/MgO (1.8 nm)/p-Si (2 μm) and (b) positive Hall resistance in Py (25 nm)/MgO (1.8 nm)/p-Si (400 nm). (c) the resistance as a function of current measured at 300 K, 100 K, 50 K, 20 K and 2 K in a Pt (25 nm)/MgO/p-Si (2 μm) sample. Inset shows voltage vs the current response. (d) The Raman spectra in strained freestanding Py(10 nm)/MgO(1.8 nm)/p-Si (2 μm) sample showing the red shift due to phonon renormalization from flexoelectronic charge carrier doping.
The transfer of the charge carrier from Py layer to p-Si layer were expected to change the Hall resistance behavior. To demonstrate, we measured the Hall resistance in two samples: Py(25 nm)/MgO(1.8 nm)/p-Si (2 µm) and Py(25 nm)/MgO/(1.8 nm)/p-Si (400 nm) as a function of magnetic field from 8 T to -8 T at 200 K as shown in Figure 3 (a) and (b), respectively. In the 2 µm p-Si sample, the Hall resistance was negative corresponding to electrons and anomalous Hall resistance was positive as shown in Figure 3 (a). Whereas in the 400 nm p-Si sample, the sign of both Hall resistance the anomalous Hall resistance were reversed as shown in Figure 3 (b). The positive Hall resistance corresponded to holes, which was expected to arise due to flexoelectronic charge transfer from Py to p-Si. Similarly, the anomalous Hall response in Py is due to intrinsic mechanism. As a consequence, the sign reversal also corresponded to the change in the type of the charge carrier from electrons to holes. This measurement provided a second proof of flexoelectronic doping. A similar electron to hole transition was also reported in Pt/MgO/p-Si sample[1].

To demonstrate the flexoelectronic doping, we, then, fabricated a Pt (25 nm)/MgO/ p-Si (2µm) thin film Hall bar sample. We measured the resistance as a function of applied longitudinal current and as a function of temperature as shown in Figure 3 (c). The resistivity of the Pt thin film was expected to be 2.57×10⁻⁷ Ω m[1] whereas the resistivity of the Si device layer was expected to be 1-5×10⁻⁵ Ω m. However, the resistance of the sample at 300 K was measured to be 2.02 Ω as compared to 9.61 Ω (using the smallest resistivity of the p-Si layer). It was noted that even if we used the bulk resistivity value for Pt (1×10⁻⁷ Ω m), the measured resistance could not be lower than 6.35 Ω. The new resistivity of the Si layer needed to simulate the observed sample resistance was
estimated to be $1.518 \times 10^{-6}$ $\Omega$ m, which could only occur due to charge carrier transfer from Pt layer to the Si layer due to strain gradient. This value of the resistivity was lower than the estimated value in p-Si (400 nm) sample as shown in Figure 2 (c,d). The melting point of Pt is significantly higher and, as a consequence, the residual stresses in the Pt layer were expected to larger, which led to larger flexoelectronic charge transfer. This reduction in resistance occurred since the mobility of charge carrier in p-Si is an order of magnitude larger than that of metals as stated earlier. In addition, the larger current should increase the resistance of sample due to self-heating instead resistance of the sample decreased. This behavior was similar to the previous reported larger spin-Hall magnetoresistance response with increased current[11]. The voltage-drop as a function of current also showed a non-linear behavior as shown in inset of Figure 3 (c). Hence, we attributed this behavior to larger strain gradient leading to larger flexoelectric doping of Si layer. This was the third experimental evidence of flexoelectronic doping of Si layer in a metal/Si bilayer structure. In the Py/p-Si (400 nm) sample, the charge carrier injection also led to exchange interactions, which led to Mott MIT. However, we did not observe Mott MIT in the Pt/p-Si (2 µm) sample even though the flexoelectronic doping levels were expected to be larger. We attributed this behavior absence of exchange interactions.

We have hypothesized that flexoelectronic doping and superposition of topological phonons were the underlying cause of topological magnetism of phonons. The flexoelectronic doping led to a net polarization in the Si layer. We posit that flexoelectronic doping may lead to the renormalization of phonons[12]. Using Raman spectroscopy, we measured the response in a freestanding Py(10 nm)/MgO (1.8 nm)/p-Si (2 µm) sample at zero mA (516.88 cm$^{-1}$) and 1 mA (515.97 cm$^{-1}$) of an applied current along with a control
p-Si sample (521.1 cm\(^{-1}\)) on the substrate. We also carried out two measurements at 5 mW and 60 mW on a control p-Si (2 µm) sample (on substrate) to verify the zone center optical phonon Raman peak and effect of laser heating. The Raman peak was measured to be 521.1 cm\(^{-1}\) and 519.5 cm\(^{-1}\), respectively at 5 mW and 60 mW, leading to heating contribution of \(\Delta \omega = 1.6 \text{ cm}^{-1}\)[13]. A red shift of 3.53 cm\(^{-1}\) (after subtracting the laser heating contribution) and phonon softening was observed in the zone center optical phonons, as shown in Figure 3 (d). The heating due to 1 mA current is expected to be \(\sim 30 \mu W\) and will not cause the red shift observed. The red shift corresponded to an average tensile strain of \(\sim 1.08\%\) [14], which could arise in the nanoscale samples[14] but not in the 2 µm thick samples [15]. Our contention was also supported by the lack of red shift in the Raman spectra from the control p-Si sample, as shown in Figure 3 (d). The full width at half maximum (FWHM) was estimated to be \(\sim 7.5 \text{ cm}^{-1}\) in both of the samples despite the Si layer being single crystal as compared to \(\sim 4.65 \text{ cm}^{-1}\) in a control prime Si wafer[13]. The charge carrier concentration in doped-Si is related to the lattice strain. The red shift in the Raman measurement was attributed to the flexoelectronic charge carrier injection from Py layer to Si layer. The excess electron and resulting polarization led to the phonon renormalization, which was also expected to be the underlying reason for topological magnetism of phonons. Previously, a similar red shift was attributed to the spin polarization[13]. However, flexoelectronic doping was believed to be mechanistic origin of red shift since it was also observed in the Pt/p-Si sample.
Figure 4. The angle dependent magnetoresistance measurement in the zy-plane (a) at 400 K as a function of magnetic field, (b) at 350 K as a function of magnetic field, (c) at 350 K as a function of current, and (d) at 300 K as a function of current and magnetic field. P and AP represent parallel and anti-parallel configuration for the Py and p-Si magnetic moments where p-Si topological magnetic moment is fixed and Py magnetic moment was aligned with the external magnetic field.

Next, we needed a direct evidence of the magnetic moment in the Si layer to verify the topological electronic magnetism of phonons. Previously, the spin-Hall effect has been reported in the 2 µm Si thin films, which was expected to arise due to phonon skew scattering. The strain gradient and flexoelectronic polarization in 400 nm Si would be larger giving rise to the large topological magnetic moment in Si. Hence, we measured
the angle dependent magnetoresistance of the sample as function of magnetic field, current and temperature as shown in Figure 3. First, we measured the angle dependent magnetoresistance in zy-plane (plane perpendicular to current direction) at 400 K as shown in Figure 4 (a). At 1 T, we measured the angle dependent behavior similar to spin-Hall magnetoresistance having $\sin^2 \theta_{zy}$ symmetry as show in Figure 3 (a)[11]. However, the symmetry of the response changed to $\cos \theta_{zy}$ for larger magnetic fields as shown in Figure 4 (a). This response was attributed to the giant magnetoresistance (GMR) for a current in-plane (CIP) geometry. The GMR was $\sim$1.22% at 14 T and 10 T magnetic fields.

Based on the symmetry, the topological magnetic moment in the Si layer was expected to be in the +z-direction and remained fixed while the Py magnetic moment always aligned with the magnetic field. This was opposite to the previous measurement reported in part 2 of this series. This also demonstrated that direction of topological magnetic moment was function of flexoelectronic effect (distribution of charge carrier) and magnitude of strain gradient. The response at 350 K was similar but the magnitude of the GMR was reduced to 1.11% at 14 T magnetic field as shown in Figure 4 (b). We also measured the response as a function of current at 350 K as shown in Figure 4 (c). The GMR response increased as a function of current at 350 K as shown in Figure 4 (c), which was attributed to the flexoelectronic doping since it would also increase interfacial scattering. Then, we measured the responses as a function of current and magnetic field at 300 K as shown in Figure 4 (d). At 300 K, the GMR response decreased to $\sim$0.51% at 10 T magnetic field. The reduction in the response due to the temperature was again attributed to the topological electronic magnetism of phonons. The reduction in temperature caused the reduction in phonon amplitude, which in turn reduced the topological electronic magnetic
moment. However, at lower field, the SMR response gave way to anisotropic magnetoresistance (AMR) response as shown in Figure 4 (d). It is noted that AMR of Py layer was always present in every measurement but was observed only when GMR and SMR responses weaken at lower temperatures. In this second step, we experimentally confirmed the topological magnetism of phonons in Si from GMR response.

![Figure 5](image)

Figure 5. (a) A schematic showing the flexoelectronic charge separation in Si later due to interfacial flexoelectric effect at the MgO interface. The Hall effect measurement in the MgO (2 nm)/p-Si (400 nm) sample (b) at 350 K for an applied magnetic field from 3 T to -3 T and (d) at 200 K from 0 to 3 T. The extracted oscillatory response (c) at 350 K and (e) at 200 K. The Hall effect measurement from 1 T to -1 T at 200 K.

While we have presented the evidence of flexoelectronic doping and topological magnetism but we needed a direct evidence of superposition of flexoelectronic doping
and topological phonons. In addition, we wanted to remove the requirement of a metallic layer for the flexoelectronic effect. We deposited 2 nm MgO on a freestanding 400 nm of p-Si sample[16]. In this case, the flexoelectric polarization of MgO layer will lead to flexoelectronic charge separation in the Si layer as shown in Figure 5 (a). In addition, the MgO layer was deposited using RF sputtering, which could lead to charge accumulation at the interface and contributed to the flexoelectronic charge separation. We, then, measured the Hall response as a function of magnetic field at 350 K and 200 K. At 350 K, the Hall measurement from 3 T to -3 T showed an oscillating behavior as shown in Figure 5 (b). It was similar to Quantum Hall effect except instead of quantization we observed sinusoidal response. In addition, the longitudinal resistance was unaffected by this oscillatory Hall response. Using a line fit, we measured the Hall resistance and extracted the oscillatory response as shown in Figure 5 (c). The Hall resistance was negative and the charge carrier concentration was estimated to be $9.7 \times 10^{18}$ cm$^{-3}$. The period of the oscillation was estimated to be ~1.2 T. At 200 K, the oscillatory Hall response was pronounced as shown in Figure 5 (d). The Hall resistance was negative and the charge carrier concentration was estimated to be $7.9 \times 10^{18}$ cm$^{-3}$. Similar to 350 K, the oscillatory response had a period of ~1.2 T as shown in Figure 5 (e). However, the oscillatory response had an opposite sign for based on direction of field sweep as shown in Figure 5 (e). The superposition of flexoelectronic charge separation with the topological phonons in Si thin film gave rise to topological magnetic moment of phonons as described by equation 5. The topological magnetism gave rise to spin-splitting of electronic band structure. The application of an external magnetic field led to Zeeman splitting in the flexoelectronic charge distribution. As a consequence, the superposition of the spin-
splitting from the topological magnetic moment and Zeeman splitting was expected to give rise to the oscillatory Hall effect. The oscillations were expected to disappear if the externally applied magnetic field was smaller than the period of oscillation. We, then, measured the Hall resistance as a function of magnetic field from 1 T to -1 T at 200 K. In this measurement, the charge carrier concentration was estimated to be \(7.34 \times 10^{18}\) cm\(^{-3}\). This measurement showed that the linear Hall response was similar for both measurements. In this new measurement, the oscillatory component was completely absent as shown in Figure 5 (f). An anomalous hysteretic behavior was also observed in the Hall response for positive magnetic field as shown in Figure 5 (f), which was expected to arise from topological magnetism. However, the mechanistic origin for this hysteretic response was unknown. This was the first experimental evidence of oscillatory Hall effect due to topological phonons. This experiment also conclusively proved out initial hypothesis of topological magnetism described by equation 5.

In conclusion, we presented the first experimental evidence of topological electronic magnetism of phonons due to flexoelectronic doping. The flexoelectronic doping arose from the charge carrier injection from the metal layer to the Si layer due to interfacial flexoelectronic effect. The flexoelectronic doping may have contribution from the mobility gradient as well. The flexoelectronic doping led to phonon renormalization that was discovered using Raman spectroscopy. The topological magnetism gave rise to giant magnetoresistance response. The superposition of the topological phonons and flexoelectronic charge separation was discovered using oscillatory Hall effect. The oscillatory Hall effect response arose due to superposition of the spin splitting due to topological magnetism of phonons and Zeeman splitting from external magnetic field. The
flexoelectronic doping and topological electronic magnetism of phonons can lead a new class of magnetoelectronic multiferroic materials for energy efficient device applications. The flexoelectronic doping can also lead a potential pathway to high temperature superconductivity.

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Supplementary: Topological phonons in an inhomogeneously strained silicon-3: Flexoelectronic doping mediated topological electronic magnetism of phonons and Mott transition

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Supplementary Table S1. The flexoelectronic charge carrier transfer from Py to Si leading to change in charge carrier concentration in each layer and resulting resistivity behavior for each current.

| I         | ρPy (Ωm) | Ratio | nPy (cm⁻³) | Reduction | nSi after flexoelectronic doping | Expected ρSi (Ωm) | Calculate ρSi (Ωm) | Error (%) |
|-----------|----------|-------|------------|-----------|---------------------------------|-------------------|--------------------|-----------|
| 10 µA     | 5.56×10⁻⁷| 1.4   | 4.89×10²²  | 1.96×10²² | 1.26×10²¹                       | 1.64×10⁻⁶         | 1.33×10⁻⁵         | 78.3      |
| 0.25 mA   | 5.89×10⁻⁷| 1.407 | 4.87×10²²  | 1.98×10²² | 1.28×10²¹                       | 1.62×10⁻⁶         | 1.15×10⁻⁵         | 79.3      |
| 0.5 mA    | 5.95×10⁻⁷| 1.493 | 4.59×10²²  | 2.26×10²² | 1.46×10²¹                       | 1.55×10⁻⁶         | 8.65×10⁻⁶         | 90.5      |
| 0.75 mA   | 5.99×10⁻⁷| 1.504 | 4.55×10²²  | 2.30×10²² | 1.48×10²¹                       | 1.53×10⁻⁶         | 6.94×10⁻⁶         | 91.8      |
The temperature dependent resistivity of the Py can be described by the following equation:

\[
\rho_{Py}^0 = 3.184 \times 10^{-7} - 4.07 \times 10^{-11} T + 2.872 \times 10^{-12} T^2 - 1.96 \times 10^{-15} T^3
\quad (S1)
\]

The residual resistance of the composite sample was larger than the resistance of Py at 5 K, which was considered residual resistance of Py only. Hence, the resistivity of Py in the composite sample at 10 µA will be \(1.4\rho_{Py}^0\) based on the residual resistance at 5 K. The column 3 represents the multiplication factor for each current dependent measurement. This factor was then used to calculate the new charge carrier concentration shown in column 4. We also estimated the reduction in charge carrier concentration (column 5) based on increase in resistivity. The number of charge carrier were then distributed to Si layer, which gave a new change carrier concentration of Si layer (column 6). Based on the column 6 data, we recalculated the resistivity of Si layer as shown in column 7. We, then subtracted the Py resistance from the overall resistance to get the resistance and resistivity response of the p-Si layer, which is second to last column in the Table S1. The difference in two resistivity based on two methods is given in the last column. This error can be attributed to the reduction in the mobility of the Si layer.

| Current (mA) | \(\rho\) (\(\Omega\)) | \(n\) (\(10^{22}\)) | \(p\) (\(\Omega\)) | \(n\) (\(10^{21}\)) | \(\rho\) (\(\Omega\)) | \(\rho\) (\(\Omega\)) |
|-------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| 1.0         | 6.02 \times 10^{-7} | 1.512           | 4.53 \times 10^{22} | 2.32 \times 10^{22} | 1.49 \times 10^{21} | 1.51 \times 10^{6} | 5.86 \times 10^{6} | 92.8 |
| 1.5         | 6.14 \times 10^{-7} | 1.54            | 4.44 \times 10^{22} | 2.40 \times 10^{22} | 1.54 \times 10^{21} | 1.46 \times 10^{6} | 4.42 \times 10^{6} | 96.1 |
| 2           | 6.21 \times 10^{-7} | 1.56            | 4.39 \times 10^{22} | 2.46 \times 10^{22} | 1.58 \times 10^{21} | 1.43 \times 10^{6} | 3.98 \times 10^{6} | 98.4 |
Supplementary Figure S1. The expected resistance behavior of Py layer as a function of temperature.

\[ \rho(T) = 3.184 \times 10^{-7} - 4.07 \times 10^{-11}T + 2.872 \times 10^{-12}T^2 - 1.96 \times 10^{-15}T^3 \]