Letter
Editors' Suggestion

Piezomagnetism in the Ising ferromagnet URhGe

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Piezomagnetism, the linear response between strain and magnetic field, is a relatively unexplored cross correlation but has promising potential as a novel probe of time-reversal symmetry breaking in various classes of materials. Interestingly, there has been no report of piezomagnetism in ferromagnets, most archetypal time-reversal symmetry-broken materials. This half-century absence of piezomagnetic ferromagnets is attributable to complications originating from multiple-domain states, as well as from changes in the magnetic point group by rotation of the magnetic moment. Here, we report characteristic V-shaped magnetostriction in the Ising itinerant ferromagnet URhGe, observed by simultaneous multiaxis strain measurements utilizing optical fiber Bragg grating sensors. This magnetostriction occurs only under fields along the *c* axis and does not scale with the square of magnetization. Such an unconventional feature indicates piezomagnetism as its origin. Our observation is owing to monodomain switching and Ising magnetization. The obtained piezomagnetic coefficients are fairly large, implying that Ising ferromagnets are promising frontiers when seeking for materials with large piezomagnetic responses.

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Cross correlations, coupling between physical quantities with orthogonal symmetry properties in systems lacking the corresponding symmetry, have been attracting much attention [1]. In particular, breaking of the most fundamental symmetries, namely the inversion and time-reversal symmetries, has been studied extensively, and novel cross-correlation phenomena such as magnetoelectric effects in multiferroics have been established [2,3]. Nevertheless, there are many unexplored cross correlations, which are worth extensive investigations both from fundamental and applicational points of view.

One such unexplored cross correlation is piezomagnetism (PZM). The PZM or piezomagnetic effect refers to the phenomenon that the strain ε of a magnetic material responds linearly to the external magnetic field H (i.e., $\varepsilon \propto H$), or its inverse effect, namely the magnetization M induced linearly by external stress σ ($M \propto \sigma$). The former is also called linear magnetostriction. These effects were first predicted in 1928 [4] and its basic theory was established in 1956 [5]. It is now understood that materials having symmetry groups without an independent time-reversal operation or those with a time-reversal operation but only in combination with lattice reflections or rotations can exhibit PZM [6]. Experimentally, PZM was first discovered in the antiferromagnets CoF_2 and MnF_2 in 1959 [7,8] following a theoretical prediction [9]. In these materials, time-reversal symmetry is lost due to the characteristic magnetic structure with up and down magnetic moments sitting respectively on different crystalline sublattices. This is in clear contrast with ordinary antiferromagnets, whose symmetry groups possess a time-reversal operation coupled with lattice translations. Recently, noncollinear antiferromagnet UO_2 is reported to exhibit hard PZM with a coercive field of as large as 18 T [10]. Some canted antiferromagnets such as α-$Cu_2V_2O_7$ [11] are also reported to exhibit PZM. More recently, PZM is attracting renewed attention as a powerful tool to detect nontrivial time-reversal symmetry breaking (TRSB) in novel states such as magnetic multipole orders [12] and altermagnetism [13,14]. Nevertheless, PZM has been only reported in several limited materials so far [6–8,10–13], and urges further investigations.

As mentioned above, PZM is allowed in systems that exhibit TRSB [5,6]. Because of this principle, we can easily predict that ferromagnets should also exhibit piezomagnetism. However, surprisingly, PZM has been observed only in antiferromagnets [6–8,10–13]. The observation of PZM in ferromagnets is perhaps hindered by complex domain configurations with various magnetization directions. In such multidomain states, bulk PZM would be canceled among domains with opposite magnetic moments. In addition, ordinary ferromagnetic (FM) magnetostriction, namely strain due to a domain configuration change, dominates [15]. Moreover, if directions of magnetic moments vary due to domain formation and/or applied magnetic fields, the magnetic point group can also change, making the detection of PZM even more complicated. Thus, most ferromagnets exhibit ordinary magnetostriction behavior approximated as $\varepsilon \propto (H - H_{coer})^2$ in magnetic fields near the coercive field H_{coer} [16]. As a

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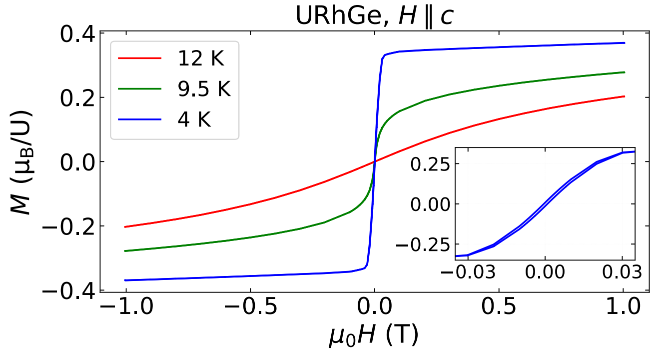

FIG. 1. Magnetization M of URhGe measured with $H \parallel c$. The inset is an enlarged view of the 4.0-K data around $H = 0$. Below T_C, $M(H)$ exhibits step-function-like behavior with a narrow hysteresis, indicating that the multidomain state is almost negligible.

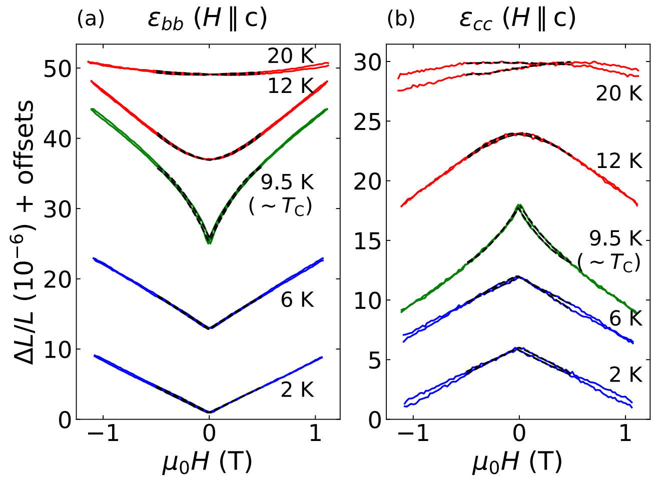

FIG. 2. Magnetostriction of URhGe in (a) ε_{bb} and (b) ε_{cc} measured under $H \parallel c$. Each curve is vertically offset for clarity. Below T_C (blue curves), V-shaped magnetostriction originating from PZM is observed in both strain components, in clear contrast to the quadratic behavior above T_C (red curves) or the nonlinear behavior with a cusp due to critical behavior at T_C (green curves). Results of fittings using Eq. (2) are shown with black dotted curves.

typical example, the FM magnetostrictive material $Tb_{0.4}Dy_{0.6}$ has been known to exhibit large and symmetric magnetostriction originating from domain changes [17]. Therefore, it is not straightforward to detect the naively expected piezomagnetism in ferromagnets.

In this Letter, we report magnetostriction in the itinerant ferromagnet URhGe measured with the multiaxis simultaneous strain measurement technique based on the fiber Bragg grating (FBG). We found unusual "V-shaped" magnetostriction in the FM phase only for specific combinations of field and strain directions. This is attributed to the ferromagnet PZM, which has been overlooked for more than a half century.

Our target material is the Ising-like itinerant ferromagnet URhGe. URhGe has the orthorhombic *Pnma* crystal structure [18]. This material exhibits ferromagnetism below the Curie temperature $T_C = 9.5$ K [18,19] and subsequently superconductivity below $T_{sc} = 0.28$ K [20]. Due to the strong spin-orbit coupling, the magnetic moment in URhGe shows strong Ising features with the easy axis along the c direction [21]. Such an Ising nature is inherited in the FM state. Recently, Mineev discussed PZM in the FM and superconducting states of URhGe and its related materials UCoGe and UGe_2 [22]. Because of the Ising magnetic anisotropy, the magnetic point group of the FM state is well defined to be $D_{2h}(C_{2h})$ irrespective of magnetic-field directions as long as the field is not too strong. For this magnetic point group, PZM is indeed allowed; the piezomagnetic effect obeys $\varepsilon_\mu = \sum_k Q_{k\mu} H_k$ with the nonvanishing piezomagnetic tensor

$$Q_{k\mu} = \begin{pmatrix} 0 & 0 & 0 & 0 & Q_{15} & 0 \\ 0 & 0 & 0 & Q_{24} & 0 & 0 \\ Q_{31} & Q_{32} & Q_{33} & 0 & 0 & 0 \end{pmatrix}, \qquad (1)$$

where ε_μ is the strain expressed using the Voigt notation (ε_1 corresponding ε_{aa}, etc.), and H_k is the k component of the magnetic field ($k = 1$, 2, and 3 corresponding to the a, b, and c directions) [6,22]. This tensor indicates that c-axis field will produce piezomagnetic linear magnetostriction in ε_{aa}, ε_{bb}, and ε_{cc}, whereas there is no PZM in these normal strains for fields along the a or b axis.

For the present study, we used high-quality single crystals of URhGe prepared with the Czochralski method. Strain ε_{bb} and ε_{cc} of this sample was simultaneously measured by using FBG sensors. The FBG is a periodic grating embedded to the core of an optical fiber and we can sensitively measure the strain from the change in the wavelength of the light reflected from an FBG pasted to the sample [23]. The strain transmission rate between the sample and FBG through glue is calibrated based on thermal-expansion measurements. To introduce light to FBGs and measure the spectra of reflected light, we used a commercial interrogator (KYOWA EFOX-1000B-4). The strain measurements were performed in a commercial cryostat [Quantum Design, Physical Property Measurement System (PPMS)], whereas magnetization M was measured with a commercial magnetometer [Quantum Design, Magnetic Property Measurement System (MPMS)]. Details of the experimental method are explained in the Supplemental Material (SM) [24].

First, in Fig. 1, we show M of the URhGe sample for H along the easy-magnetization axis (c axis) measured at various temperatures. In the FM state below $T_C \sim 9.5$ K, $M(H)$ exhibits a steplike change around $H = 0$ with a very narrow hysteresis width. Note that the saturated moment is about $0.4\mu_B$/U, which is much smaller than the value expected for U $5f^2$ or the $5f^3$ configuration ($\sim 3\mu_B$/U), indicating weak itinerant ferromagnetism. The steplike change indicates that all magnetic moments flip simultaneously without forming FM domains. Thus, this compound is free from complicated phenomena originating from multidomain configurations. This is perhaps due to the large energy cost of domain formations: In Ising ferromagnets, the stray field cannot be effectively reduced by forming multidomain states. Moreover, domain walls are not trivial, since typical FM domain walls such as Bloch or Néel types accompany magnetization rotations, which are prohibited in Ising ferromagnets.

Next, in Fig. 2, we show our main result, namely the magnetostriction measured under $H \parallel c$ at various temperatures. Above T_C (red curves), both ε_{bb} and ε_{cc} exhibit quadratic

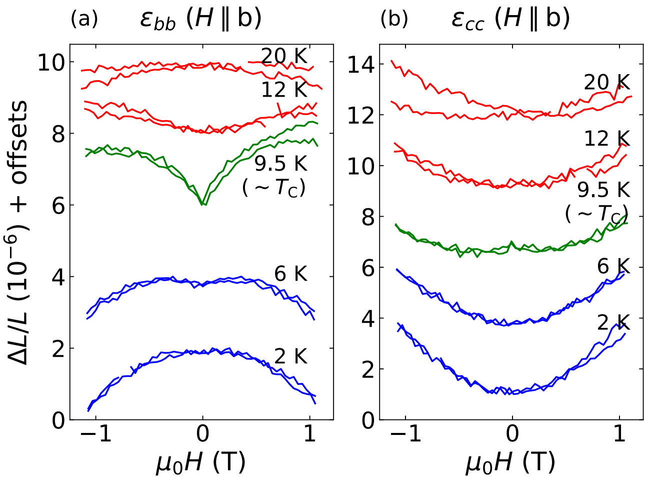

FIG. 3. Magnetostriction of URhGe in (a) ε_{bb} and (b) ε_{cc} measured under $H \parallel b$. Each curve is vertically offset for clarity. V-shaped magnetostriction was not observed below T_C (blue curves), whereas the behavior above T_C (red curves) and at T_C (green curves) qualitatively resembles those observed under $H \parallel c$ (Fig. 2).

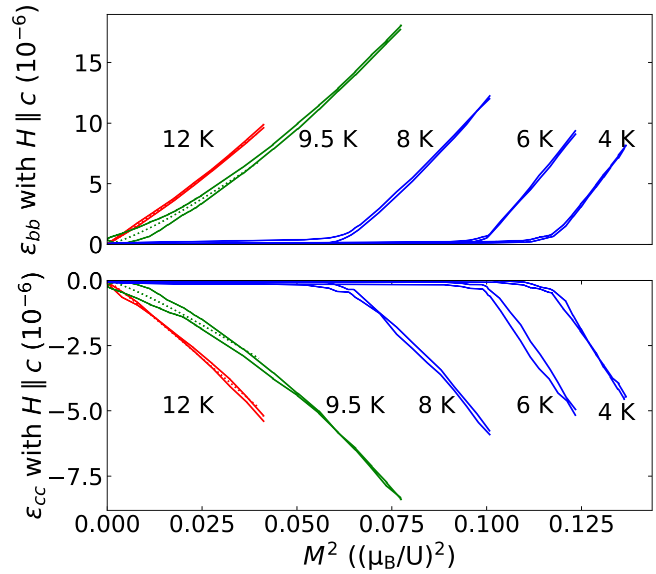

FIG. 4. Relation between ε_{cc} and M^2 under $H \parallel c$ at various temperatures deduced from the data in Figs. 1–3. The strain is almost proportional to M^2 above T_C (red solid curve) as demonstrated by the linear fit shown with the red dotted line. At T_C (green curve), the curvature is nonzero, but the power-law relation still holds between strain and magnetization. In clear contrast, there is no simple power-law relation between them in the FM phase (blue curves).

behavior around zero field. This is ordinary behavior for paramagnets; if M is proportional to H, the ordinary magnetostriction should obey the relation $\varepsilon \propto M^2 \propto H^2$.

At $T_\mathrm{C} = 9.5$ K (green curves), ε_{bb} and ε_{cc} show sharp kinks at $H = 0$, followed by nonzero curvatures at finite fields. At first glance, this looks anomalous but is mostly attributable to the ordinary magnetostriction $\varepsilon \propto M^2$ together with the critical behavior in M at T_C: Mean-field theories predict $M \propto H^{1/3}$ at the transition temperature [25], hence ε is expected to show nonlinear $H^{2/3}$ behavior. Indeed, such cusplike magnetostriction at T_C has been reported in other ferromagnets such as UIr [26] and $\mathrm{TbCo_2Mn}_x$ [27]. The behavior at T_C will be discussed again later.

Interestingly, in the FM state (blue curves), $\varepsilon_{bb}(H)$ and $\varepsilon_{cc}(H)$ curves are strikingly V shaped, with robust H-linear dependences in both $H > 0$ and $H < 0$ regions. As a consequence, ε is proportional to $|H|$. This behavior remains down to 2 K, the lowest temperature of the present study. Compared with the steplike $M(H)$ curve in the FM phase (Fig. 1), it is clear that the $\varepsilon \propto |H|$ dependence cannot be described by ordinary magnetostriction $\varepsilon \propto M^2$, as demonstrated in Figs. S6 and S7 [24]. Below, we will discuss other features to conclude that this V-shaped magnetostriction originates from the PZM in URhGe.

Here, we comment on the difference in the sign of magnetostriction. The difference in the sign of the magnetostriction reflects the anisotropic thermal expansion at zero field (Fig. S2): When the magnetic moment grows as temperature decreases, the b axis tends to expand and the c axis tends to shrink. The application of the c-axis magnetic field, bringing the sample closer to or deeper in the FM state, causes the same effect, leading to the observed sign anisotropy in the magnetostriction under $H \parallel c$. This tendency that a shorter b axis disfavors the FM order is consistent with the fact that T_C decreases under uniaxial pressure along the b axis [28]. Moreover, this uniaxial-pressure effect is known to be consistent with the negative jump in the b-axis thermal-expansion coefficient at T_C [29] through the Ehrenfest relation. These tendencies are naively consistent with the magnetic dipole interactions repelling each other in the transverse direction, but the detailed microscopic mechanism is not yet known.

We first examine the anisotropy of the V-shaped magnetostriction. We show in Fig. 3 the magnetostriction measured under $H \parallel b$. The strain above T_C is weak and quadratic against the magnetic fields. At $T = T_\mathrm{C}$, the strain showed a cusp at $H = 0$, which is similar to those observed in $H \parallel c$ and is attributable to the critical behavior. For $T < T_\mathrm{C}$, the $\varepsilon(H)$ curves become quadratic again, in clear contrast to the V-shaped curves observed in $H \parallel c$. This quadratic behavior is consistent with the previous report of ε_{bb} measured under $H \parallel b$ [30]. Thus, the V-shaped magnetostriction has strong anisotropy depending on the field direction. This anisotropy is consistent with the PZM tensor shown in Eq. (1): For URhGe, Q_{21}, Q_{22}, and Q_{23} are zero, meaning that no PZM should occur in ε_{aa}, ε_{bb}, or ε_{cc} under $H \parallel b$.

Second, we compare ε and M in more detail. For ordinary magnetostriction, the empirical relation $\varepsilon \propto M^2$ often holds in the limit of small M. We thus plot ε_{bb} and ε_{cc} as functions of M^2 in Fig. 4. One can clearly see that, above T_C, the strain is nearly proportional to M^2. At T_C, where the cusps in $\varepsilon(H)$ curves at $H = 0$ are observed, the strain versus M^2 curves acquire nonzero curvature, but maintain smooth relations close to linear. Indeed, when we fit the curves with $\varepsilon \propto M^{2\alpha}$ using the exponent α as the fitting parameter, we obtain $\alpha = 1.10$ for $T = 12$ K and 1.27 for $T = 9.5$ K, both being close to unity. This result manifests that the strain in URhGe above and at T_C is attributed mainly to the ordinary magnetostriction. In contrast, the $\varepsilon(M^2)$ curve at 4.0 K does not show a power-law relation. Thus, the V-shaped magnetostriction does not have the conventional relation to the magnetization.

Now we explain that the observed V-shaped magnetostriction is indeed well attributable to PZM. Generally, in magnets showing TRSB, two magnetic structures connected by the time-reversal operation exhibit opposite signs of piezomagnetic coefficients, respectively. Thus, in typical piezomagnetic materials having the piezomagnetic coefficient Q for one of the magnetic structures, the strain obeys $\varepsilon = QH$ as long as the magnetic structure is kept, while the opposite behavior $\varepsilon = -QH$ emerges when the magnetic structure is reversed by a strong magnetic field exceeding H_{coer}. This results in butterflylike $\varepsilon(H)$ curves [10,12]. In URhGe under $H \parallel c$, H_{coer} is nearly zero, as demonstrated in the steplike $M(H)$ curve (Fig. 1). Zero coercive field changes the butterflylike $\varepsilon(H)$ curve to the V-shaped curve, as observed in URhGe.

These analyses and considerations confirm that the V-shaped magnetostriction observed under $H \parallel c$ in URhGe originates from PZM. It is interesting that, although ferromagnets are most archetypal examples exhibiting spontaneous TRSB, the required feature to realize PZM as theoretically established in 1956 [5], ferromagnetic PZM has been overlooked for more than 60 years. The near absence of a multidomain state near $H = 0$ and the strong Ising nature fixing the magnetic point group irrespective of the field directions are the keys to avoid various complications characteristic of ferromagnets.

To investigate the temperature evolution of the PZM, we fitted the data with the function

$$\varepsilon(H) = \varepsilon_0 + a_2(\mu_0 H)^2 + a_{\mathrm{abs}}|\mu_0 H| \qquad (2)$$

in the field range $-0.5\ \mathrm{T} \leqslant \mu_0 H \leqslant +0.5\ \mathrm{T}$. This fitting range was chosen to achieve both accuracy and stability of the fits in the whole temperature range, as discussed in detail in SM [24]. As shown with the dotted curves in Fig. 2, the fittings are successful for all data sets. The temperature dependence of a_2 and a_{abs} are shown in Fig. 5. Above T_{C}, a_2 is dominant,

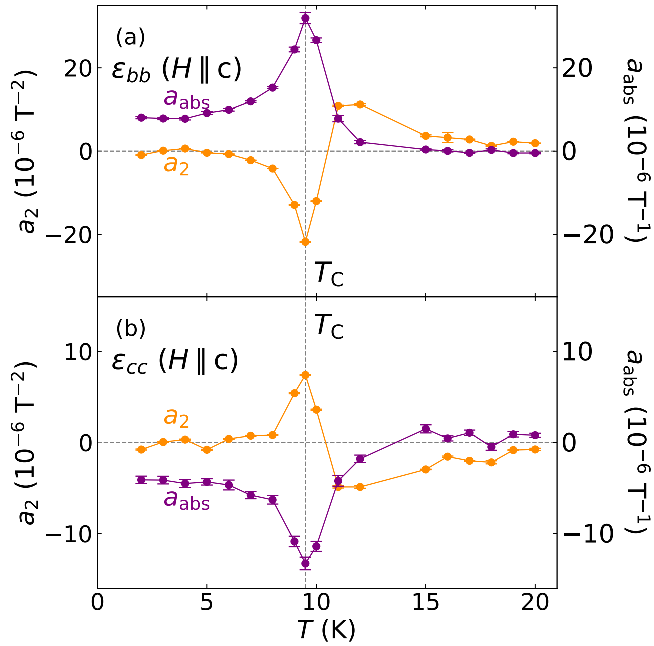

FIG. 5. Temperature dependence of the fitting coefficients a_2 and a_{abs} [Eq. (2)] for the magnetostrictions (a) ε_{bb} and (b) ε_{cc} measured under $H \parallel c$. The vertical dotted line indicates T_{C}. The dominant magnetic field dependence of the strain changes from $\varepsilon \sim H^2$ (ordinary magnetostriction) above T_{C} to $\varepsilon \sim |H|$ (PZM) below T_{C}. The singular behaviors at T_{C} are attributable to the critical nonlinear behavior in $\varepsilon(H)$ as discussed in the text.

TABLE I. Properties of known piezomagnetic materials [6–8,10–13]. The values with * were obtained by using strain-induced magnetization measurements, while the others by linear-magnetostriction measurements. Q_{poly} was obtained using polycrystalline samples. In the column of magnetization, AFM refers to an antiferromagnet, MOO a magnetic-octupole ordered material, ALM an altermagnet, and FM a ferromagnet.

Materials	$Q_{k\mu}$ (10^{-6} T^{-1})	T (K)	Magnetism	Refs.
CoF_2	$Q_{14} = \pm 21^*$	20	AFM	[8]
	$Q_{36} = \pm 9.8$	4		[31]
MnF_2	$Q_{14} = \pm 0.2^*$	20	AFM	[8]
	$Q_{14} = \pm 0.07^*$	60		[32]
α-Fe_2O_3	$Q_{22} = \pm 1.9$	78	AFM	[33]
	$Q_{22} = \pm 3.2^*$	77		[34]
	$Q_{22} = \pm 1.3$	100		[35]
	$Q_{23} = \pm 2.5^*$	292	Canted AFM	[33]
$YFeO_3$	$Q_{15} = \pm 1.7$	6	AFM	[36]
$YCrO_3$	$Q_{15} \simeq \pm 1$	6	AFM	[37]
$DyFeO_3$	$Q_{36} = \pm 6.0$	6	AFM	[38]
α-$Cu_2V_2O_7$	$Q_{\mathrm{poly}} = \pm 0.077$	5	Canted AFM	[11]
UO_2	$Q_{14} = \pm 10.5$	2.5	AFM	[10]
MnTe	$Q_{\mathrm{ave}} = 0.0050^*$	250	AFM/ALM	[13]
Mn_3Sn	$Q_{11} = \pm 4.4^*$	300	AFM/MOO	[39]
	$Q_{11} = \pm 14.6$			[12]
URhGe	$Q_{32} = +8.0$	2	FM	This work
	$Q_{33} = -4.1$			

whereas a_{abs} is within the noise level. The peaks at T_{C} are attributable to the critical behavior in $\varepsilon(H)$ discussed above: When we are forced to fit the cusplike $\varepsilon(H)$ curve using Eq. (2), we mathematically need a large $a_{\mathrm{abs}}|H|$ term and similarly large a_2H^2 term with the opposite sign. Much below T_{C}, a_{abs} becomes dominant and a_2 is nearly zero, as expected from the V-shaped magnetostriction. These results quantitatively show that the behavior of strain drastically changes from the ordinary magnetostriction $\varepsilon \propto H^2$ above T_{C} to the piezomagnetic response $\varepsilon \propto |H|$ in the FM phase. In contrast, results of the same analysis for $H \parallel b$ (Fig. S8 in SM [24]) show that a_{abs} is negligible both at $T \ll T_{\mathrm{C}}$ and $T > T_{\mathrm{C}}$, supporting again the absence of PZM in this field direction.

The fitting coefficient a_{abs} in the FM phase is equivalent to the corresponding piezomagnetic-tensor component. Taking the lowest-temperature values in the FM state, we obtain $Q_{32} = 8.0 \times 10^{-6}\ \mathrm{T}^{-1}$ and $Q_{33} = -4.1 \times 10^{-6}\ \mathrm{T}^{-1}$ for URhGe. These values are compared with results of other piezomagnets. As listed in Table I, most of the known piezomagnets exhibit $|Q_{k\mu}|$ of less than $2 \times 10^{-6}\ \mathrm{T}^{-1}$, whereas recently found piezomagnets such as UO_2 and Mn_3Sn exhibit $|Q_{k\mu}|$ exceeding $10 \times 10^{-6}\ \mathrm{T}^{-1}$ [10,12]. The observed PZM components of URhGe reach around 50% of these values. This comparison implies that ferromagnets with appropriate conditions are candidate materials realizing large piezomagnetic coefficients. We comment that the signs of Q is well defined in our case due to the very soft magnetism, whereas other piezomagnets exhibit PZM of both signs depending on magnetic structures.

One open question is the microscopic mechanism of the observed large PZM in URhGe. Naively, sufficient magnetolattice coupling is necessary to induce PZM. Such magnetolattice coupling should originate from spin magnetic moments with strong spin-orbit coupling (SOC) and/or orbital magnetic moments. Uranium compounds, exhibiting strong SOC and thus angular-momentum coupling between spin and orbitals, should satisfy both criteria. The Ising magnetic nature in URhGe indeed originates from SOC [40]. More recently, it is revealed that the nonsymmorphic crystalline structure, resulting from the zigzag uranium chain, leads to an anisotropic pseudospin texture pointing perpendicularly to the Brillouin-zone boundaries under SOC [41]. Such a pseudospin texture may explain microscopic origins of the Ising ferromagnetism in URhGe, and may further provide bases toward clarifying the mechanism of PZM.

To summarize, we revealed the piezomagnetism (PZM) in the itinerant Ising ferromagnet URhGe. The observed piezomagnetic coefficients range around 50% of the largest ones ever known. This work demonstrates that Ising ferromagnets without multidomain states can be good candidates when seeking for materials with large piezomagnetic responses, which can be utilized for sensors or actuators. This finding would stimulate further experimental and theoretical studies toward the understanding of microscopic mechanisms of PZM, which can be a probe of TRSB phenomena occurring in various materials.

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