Metastable magnetization plateaus in the S=1 organic spin ladder BIP-TENO induced by a microsecond-pulsed megagauss field

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The entire magnetization process in the two-leg organic spin ladder BIP-TENO is studied with an ultrahigh magnetic field of up to 150 T. Nontrivial multiple magnetization plateaus are observed, indicating there exist strong quantum correlations between spins. In addition to the previously reported 1/4 plateau, another plateau appears at 1/3 magnetization only when spins are decoupled to the lattice degree of freedom in the adiabatic limit. Spontaneous symmetry breaking is expected to occur with the *spin-lattice decoupling* induced by a fast evolution of external magnetic fields in the range of μs. Under the adiabatic condition, five fractional (1/4, 1/3, 1/2, 2/3, and 3/4) magnetization plateaus are likely to appear with an assumption that the magnetization saturates at around 160 T.

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I. INTRODUCTION

A spin system in a crystal is an ideal playground to study quantum statistics and rich quantum states such as the exotic spin liquid [1], magnon Bose-Einstein condensate [2], and magnetization plateaus due to the crystallization of triplons [3]. Among several spin systems, the spin ladder has been attracting great attention in terms of the intermediate dimensionality between one and two dimensions [4].

The two-leg spin-ladder compound 3,3′, 5, 5′-tetrakis(*N-tert*-butylaminoxyl)biphenyl: $C_{28}H_{42}N_4O_4$ (BIP-TENO) was synthesized as the first $S = 1$ two-leg spin ladder, where S is the spin quantum number [5]. $S = 1$ spin ladders would exhibit a unique property different from S =1/2 ladders because the ground state is expected to be the gapped Haldane state when the rung exchange interaction is extremely small. Since the dimer gapped state is expected when the leg exchange interaction is negligible, the phase transition between the two different gapped phases is expected to occur at a critical gapless point [6]. In BIP-TENO, the nontrivial 1/4 magnetization plateau was observed in the magnetic field range from 42 to 66 T and theoretically analyzed [7]. To explain the 1/4 plateau, the authors of Ref. [7] introduced the second-nearest-neighbor (diagonal direction) and third-nearest-neighbor (leg direction) exchange interactions and found that the third-nearest-neighbor exchange interaction is more essential for the appearance of the 1/4 plateau. They obtained the magnetization by a numerical diagonalization. Although further successive phase transitions to the 1/2 and 3/4 plateau states are theoretically predicted [7], they were never experimentally observed because the required magnetic field is as strong as 150 T, which is only available employing a destructive manner [8].

In the present study, the entire magnetization process of BIP-TENO is investigated in ultrahigh magnetic fields of up to 150 T using the single-turn coil that is one of the destructive means for ultrahigh magnetic field generation. It is unveiled that 1/3 and 2/3 plateaus appear in addition to the previously discussed 1/4, 1/2, and 3/4 plateaus. Moreover, the 1/3 and 2/3 plateaus are found to appear only when the magnetic field is swept in a microsecond timescale. The adiabatic condition is likely to be necessary for its observation, suggesting that the newly observed plateaus are metastable states. It would also be worth noting that the recent theoretical study [9] suggested that the 1/3 and 2/3 plateaus can also appear. The authors developed a solvable model with the condition that the exchange interactions of leg and diagonal directions are of the same magnitude, and introduced the second-nearest-neighbor interactions in the leg direction as well as in the diagonal direction. They discovered that when the magnitude of the leg exchange interaction is half of that of the rung exchange interaction, introducing the second-nearest-neighbor exchange interactions in both leg and diagonal directions can simultaneously give the multiples of 1/4 and multiples of 1/6 plateaus. Their finding is that, even by introducing further neighbor interaction in both leg and diagonal

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directions, the exactly solvable model can be constructed and the 1/3 plateau is predicted to appear in the $S = 1$ two-leg spin ladder.

Both theoretical studies [7,9] explained the appearance of the 1/4 plateau, which is consistent with the experimental results. Although the recent theory [9] indicated that the 1/3 plateau can appear, the absence of the 1/6 and 5/6 plateaus in the experimentally obtained magnetization curve in the present work may suggest that further theoretical studies are required.

In the present study, magnetostriction is found to play an important role in understanding the transient phenomenon related to the metastable states. It is found that a lattice contraction occurs when magnetic fields are applied rather slowly in the millisecond (ms) timescale, while no magnetostriction takes place if magnetic fields are swept faster than approximately 1 ms. The fast μs-long fields only control spins without movement of the lattice nor molecules, and novel plateaus can appear. In a magnetic field applied slowly, the lattice contraction can stabilize the 1/4 plateau and may prevent realizing the 1/3 plateau phase as a result of modification of the exchange interactions due to the lattice contraction. To see how the lattice contraction affects the stability of the 1/4 plateau, the theoretical investigation focusing on the 1/4 plateau is done with the density matrix renormalization group (DMRG) method. The enlargement of the particular exchange interactions due to the contraction of the lattice can qualitatively explain the stabilization of the 1/4 plateau.

II. EXPERIMENTS

The single crystals of the 3,3′, 5, 5′-tetrakis(*N-tert*-butylaminoxyl)biphenyl: $C_{28}H_{42}N_4O_4$ called BIP-TENO were synthesized by the method reported previously [5]. The typical dimensions of the crystal used are $2 \sim 4 \times 0.5 \times 0.2$ mm^3 and the direction of the long side is parallel to the *c*-axis. Three or four single crystals were used for the magnetization measurement to obtain the larger signal. In the pulsed-field experiments, magnetic fields (B) were applied parallel to the *c*-axis of the crystal that is parallel to the leg-direction of the spin ladder.

The ultrahigh magnetic fields were generated using the single-turn coil installed in The Institute for Solid State Physics, The University of Tokyo [8]. The magnetization was measured using a self-compensated parallel-type pickup coil [10]. The sample temperature was lowered by directly immersing the sample in liquid helium or by putting the sample in flowing cold helium gas.

The magnetocaloric effect was measured with a nondestructive 36-ms-duration pulsed magnet [11,12]. The magnetostriction measurements were conducted using a fiber Bragg grating (FBG) and the optical filter method was utilized [13,14]. For the FBG experiment, a miniature pulsed magnet consisting of three wire-wound coils was used for the generation of magnetic fields of up to 20 T with different pulse durations [15]. The duration time of the magnetic field is from 0.2 to 2.2 ms using a capacitor of 0.8 or 1.6 mF.

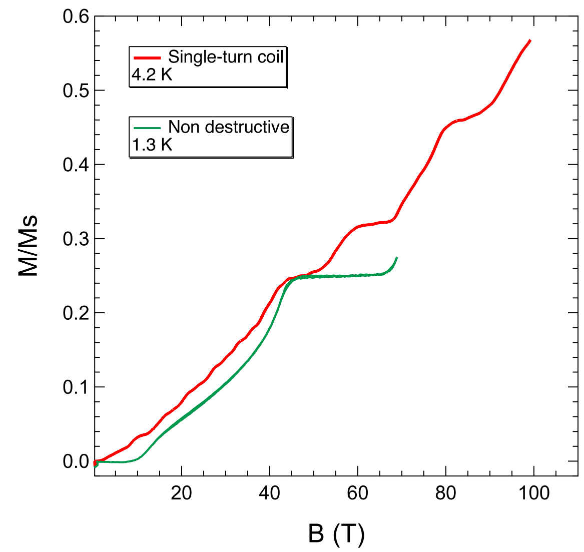

FIG. 1. Magnetization process of a single crystal of BIP-TENO measured with the μs single-turn coil (red curve). The green curve shows the magnetization process of BIP-TENO measured in a nondestructive pulsed magnetic field with a duration of 5 ms [7].

III. RESULTS OF THE EXPERIMENTS

Figure 1 shows the magnetization process measured at 4.2 K with the single-turn coil up to approximately 100 T (a red curve). Two typical waveforms of the pulsed magnetic field generated by the single-turn coil method are shown in Fig. 2. In Fig. 1, the previously reported magnetization curve at 1.3 K [7] is shown together for comparison (a green curve). A nondestructive magnet with 5-ms duration was used in this measurement.

As shown in Fig. 1, the gapped feature is seen in the green curve as almost no magnetization at low magnetic fields below around 10 T, while the magnetization gradually increases in the corresponding low field region in the red curve. This

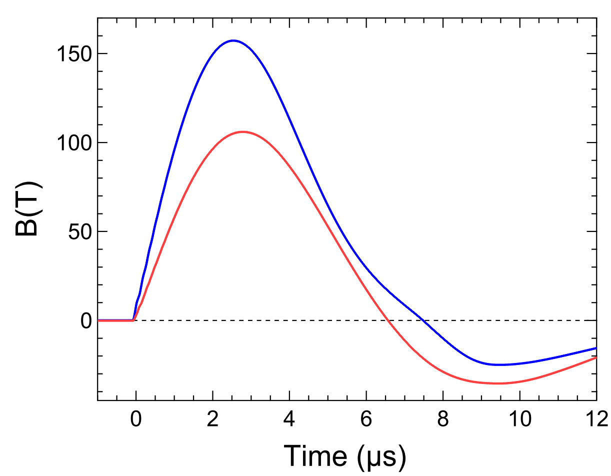

FIG. 2. Waveforms of the pulsed magnetic fields generated by means of the single-turn coil. The vertical-type (red curve) and horizontal-type (blue curve) single-turn coil field generators were utilized for generations of fields of up to 105 and 160 T, respectively [8].

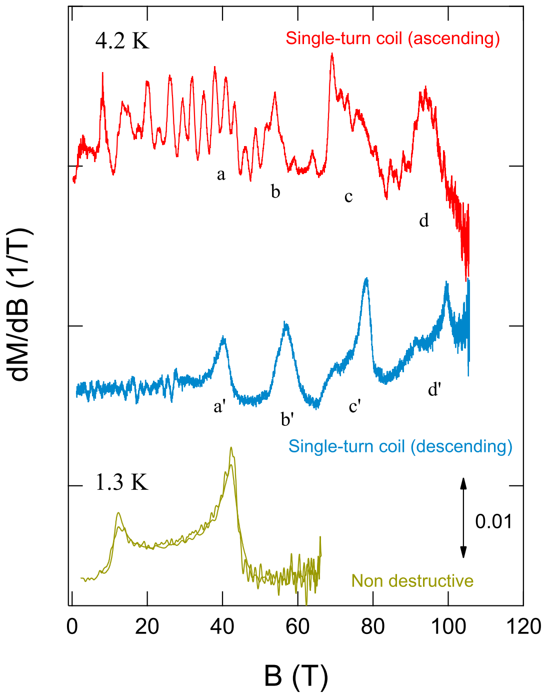

FIG. 3. dM/dB curves in the field-ascending (red) and field-descending (blue) processes are shown for the single-turn coil experiment up to 105 T. dM/dB curve in the nondestructive 5-ms-pulsed magnetic field is shown with dark yellow line.

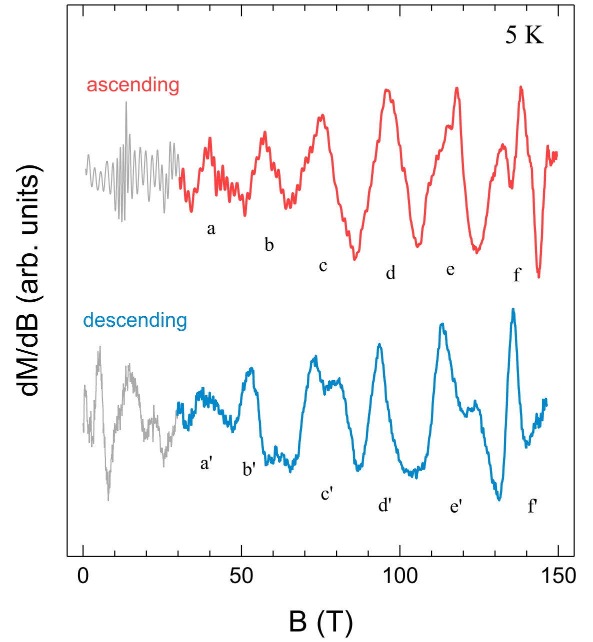

FIG. 4. dM/dB curves in the field-ascending (red) and field-descending (blue) processes are shown for the single-turn coil experiment up to 150 T. The background noise was subtracted from the original data. The initial part of the signals are shown with grey color, indicating the observed signal structures are not intrinsic because they are disturbed by the starting discharge noise of the field generation.

difference can be understood as the thermal broadening effect; the spin is thermally excited even at 4.2 K in low magnetic fields. Although the magnetic field is generated up to 105 T, the results obtained in fields over 100 T were found to be affected by the background noise rather considerably and to be less reliable. Hence, the magnetization curve is plotted up to the field slightly below 100 T. We also discuss this point when the dM/dB signal is plotted.

The most striking feature seen in the red curve is that the 1/4 plateau is terminated by the appearance of another plateau at around 50 T at which the 1/4 plateau remains in the green curve. This significant difference between the red and green curves can be due to the duration time of the magnetic field. The three orders of magnitude difference of the field duration (ms versus μs) can give rise to the significant difference of the magnetization process. In the red curve, the M/M_s of the newly observed two successive plateaus at 60–70 and 80–90 T are 0.32 ± 0.01 and 0.46 ± 0.05, respectively, where M is the magnetization and M_s is the saturation magnetization. Hence, these two successive plateaus can be assigned to the 1/3 and 1/2 magnetization plateaus, respectively. The larger error in the higher fields is due to a reduction of the sensitivity of the magnetization measurement near the top of the magnetic field pulse: dM/dt that is proportional to the signal becomes zero at the maximum field.

In Fig. 3, the dM/dB is plotted as a function of B. The red and blue curves show the dM/dB for the field-ascending and field-descending processes of the single-turn coil experiment, respectively. The dark-yellow curve shows the dM/dB for the 5-ms nondestructive pulsed fields [7]; the field-ascending and field-descending processes are nearly overlapped in the millisecond time resolution. Here, we should note that the magnetization curve up to 100 T shown in Fig. 1 is obtained by taking an average of the field-ascending and field-descending dM/dB signals. The background signal that seems to depend on the field sweeping direction is found to be canceled when taking the average. It is worth mentioning that the cancellation is not well done when the field reaches close to the top of the pulse. The background noise of the dM/dB signal becomes more significant in fields exceeding 100 T. Since the dM/dB is deduced from $(dM/dt)/(dB/dt)$ and both dB/dt and dB/dt become small near the top of the pulse, the measurement sensitivity becomes less and the reliability of the result becomes low when the field reaches close to the top of the pulse. Hence, we plot the magnetization up to the field slightly lower than 100 T in Fig. 1.

We put labels a, b, c, and d for each peak structure found in the field-ascending dM/dB curve (red curve) in Fig. 3. In the same manner, a′, b′, c′, and d′ are put for the field-descending dM/dB curve (blue curve). The peaks from a to d likely correspond to the a′, b′, c′, and d′ peaks.

In Fig. 4, the dM/dB obtained in the experiment at 5 K with a higher magnetic field of up to 150 T are plotted as a function of B for field-ascending and field-descending processes, respectively. Because the dM/dB is found to be affected by a background signal more sensitively at near the top of the magnetic field, we only plot the results up to 150 T, although the field is generated up to 160 T as shown in Fig. 2. The dM/dB signal is obtained after subtracting the background noise from the raw data. Here, the background noise is represented as a polynomial function because it is propor-

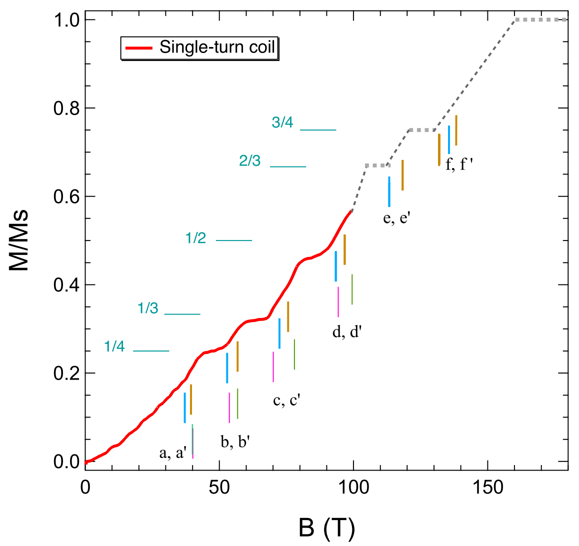

FIG. 5. Magnetization curve observed and its extension schematically indicated with dashed lines. The short vertical lines indicate the dM/dB peak positions. For the 150 T experiment, the light-blue and dark-yellow lines represent the results of the field-ascending and field-descending measurements, respectively. For the 100 T experiment, the green and pink lines represent the results of the field-ascending and field-descending measurements, respectively. The horizontal lines denote the normalized magnetization at each plateau expected.

tional to the dB/dt and cannot be canceled out even taking the average of the field-ascending and field-descending processes for the 150 T experiment. The difference in the background noises of the 100 T and 150 T experiments is mainly due to the size difference of the single-turn coil. The diameter of the coil is 14 mm for the 100 T experiment and it is 12 mm for the 150 T experiment. The spatial homogeneity of the magnetic field and its time variation depend on the size of the coil. The smaller coil gives less homogeneity. Because the background signal of the magnetization measurement becomes large when the magnetic field homogeneity becomes less, the background noise becomes more significant in the 150 T experiment using a 12-mm coil.

Two peaks at around 120 T (e and e$'$) and 135 T (f and f$'$) are additionally observed above 100 T. Although the correct estimation of magnetization curve is difficult at this stage owing to the large background signal, we could estimate the peak position from the present dM/dB curve.

Figure 5 shows the magnetization process up to 100 T and the positions of the dM/dB peaks obtained from data shown in Figs. 3 and 4. It is found that, although the positions are slightly different even for peaks with the same label such as a or b$'$ depending on the maximum field of 100 or 150 T, each x and x$'$ (x = a, b, c, and d) peaks seem to be located at a particular field range where a slope starts in the magnetization curve.

Here we try to evaluate the higher-field magnetization curve over 100 T by a rough estimation of the magnetization behavior using the field positions of the peaks observed in the dM/dB curves. In the first place, looking at the peak positions from b(b$'$) to d(d$'$) in Fig. 5, these peaks are found to be located at the beginning of the slope of the magnetization after the plateaus. Hence, it would not be unreasonable to assume that the e(e$'$) and f(f$'$) located at the beginning of the slopes of the magnetization process in a higher field region than 100 T. Second, because the plateaus at $M/M_s = 2/3$ and $3/4$ are predicted to appear in the higher magnetic fields adding to the observed $1/4$, $1/3$, and $1/2$ plateaus [7,9], we can assume that these potential magnetization plateaus would be followed by each magnetization slope. Namely, the most plausible plateaus associated with the e(e$'$) and f(f$'$) peaks are the $2/3$ and $3/4$ plateaus, respectively.

Third, we simply evaluate the magnetization process by drawing magnetization curves with linear slope such that the dM/dB peaks of e, e$'$ and those of f, f$'$ located at the beginning of the slope of the magnetization curve after $2/3$ and $3/4$ plateaus, respectively. The length of the plateaus is taken to be 9.5 T, which is approximately as large as those of $1/4$ and $1/3$ plateaus. (The lengths of the $1/4$ and $1/3$ plateaus were observed to be 9 and 10 T, respectively.) Finally, we assume that the saturation of the magnetization occurs at 160 T as proposed by the theoretical estimation [7]. Based on these assumptions, we estimate the possible magnetization curve above 100 T as shown in Fig. 5 by the grey dashed curve and propose the entire magnetization process up to the saturation field.

IV. ORIGIN OF FIELD SWEEP RATE DEPENDENCE OF THE MAGNETIZATION PROCESS

The magnetization process obtained with the single-turn coil (short pulsed durations of several microseconds) are found to exhibit the $1/3$ plateau that was not observed in the relatively slow measurement of a millisecond timescale. Moreover, its counterpart $2/3$ plateau as well as $1/2$ and $3/4$ plateaus are likely to appear in the entire magnetization curve as shown in Fig. 5. We believe that the appearance of $1/3$ and $2/3$ plateaus are due to the rapid field sweep in the microsecond timescale. We would like to discuss the possible origins of this unusual phenomenon.

A decrease in the sample temperature due to the magnetocaloric effect in a pulsed magnetic field is one of the possible origins. Because of the magnetocaloric effect, sample temperature changes when magnetization changes in adiabatic conditions. When the pulse duration is several microseconds, almost no thermal relaxation to the surrounding thermal bath occurs due to the short time allowed for the present magnetization experiment. Therefore, the sample temperature inevitably changes during the magnetization process for the single-turn coil experiment, and a possible significant temperature lowering due to the magnetocaloric effect may induce the $1/3$ plateau at around 50 T. A related phenomenon is observed in the Sastry-Sutherland magnet $SrCu_2(BO_3)_2$ [16–18]. Although the $1/8$ plateau is observable at a temperature lower than 0.5 K when a DC magnet was used [16], it was observed even at 2.1 K when the single-turn coil was employed [18], suggesting the lowering of temperature in the sample by the fast sweep rate of the magnetic field.

To elucidate the origin of $1/3$ magnetization plateau, the magnetocaloric effect (MCE) was measured on BIP-TENO

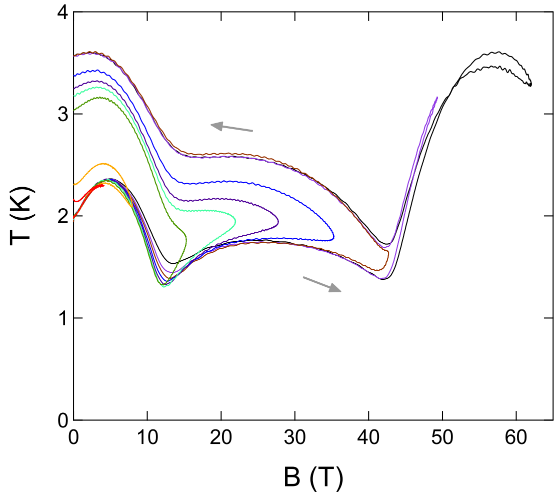

FIG. 6. Magnetocaloric effect at the initial temperature of 2 K with several maximum field strengths.

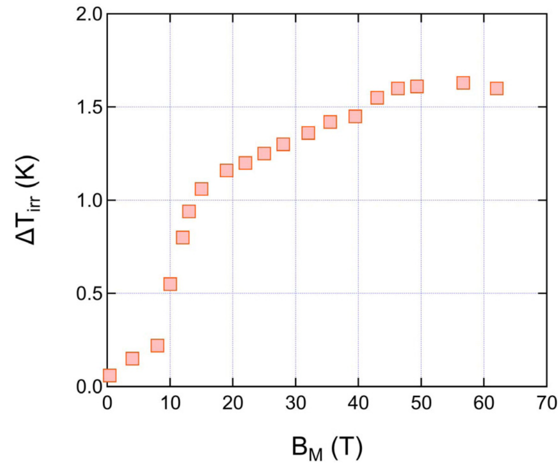

FIG. 7. Temperature difference ΔT_{irr} between the initial and final temperatures measured in the MCE experiment. ΔT_{irr} depends on a maximum value of a pulsed magnetic field B_M.

in a nondestructive pulsed magnet with a pulse duration of 36 ms. Figure 6 shows the MCE traces $T(B)$ measured at the initial temperature of 2 K with several maximum field strengths. The sample temperature is measured as a function of a magnetic field. The eight curves shown in different colors are the results of the measurements up to different maximum fields.

The sample is thermally quasi-isolated from the surroundings [11,12] so that there is no heat leak from or to the surrounding as like as the magnetization measurements with single-turn coils. Because the MCE measurement is conducted rather slowly in the millisecond time scale, the spin and lattice are expected to be well thermally coupled. The sample temperature clearly changes as a function of magnetic fields and exhibits sharp dip structures at around 12 and 42 T. These sharp dip structures in MCE manifest themselves as the peak of entropy surface $S(B, T)$ at the phase boundary [19]. The dome-like structure of MCE around 55 T, corresponding to a dip structure in $S(B, T)$, suggests the spin-gap opening on the 1/4 plateau [20]. Albeit in the magnetization process taken in the microsecond pulse, there is no signature of a phase transition between 42 and 62 T in the MCE data that is taken in pulsed-field of 36-ms-pulse duration. We emphasize that both measurements are carried out with no heat exchange with the surrounding heat baths. The difference is that the MCE measurement allows the spin system to thermalize to the lattice, but the present magnetization measurement might not have enough time to exchange heat between the lattice and the spin systems due to the limited measurement timescale. Furthermore, the minimum temperature achieved in the present MCE measurement (1.2 K) is compatible with the measurement temperature (1.3 K) of the magnetization process in millisecond pulsed fields [7], suggesting that the MCE cannot explain the observation of the 1/3 plateau at around 50 T in this temperature region. We, therefore, judge that the 1/3 plateau is observed in the present magnetization experiment due to its rapid field sweep rate.

It should be noted that there is significant irreversibility in the magnetocaloric effect. The two arrows in Fig. 6 indicate the field ascending and descending process, and it is found that the final temperature after applying a pulsed field is higher than the initial temperature. One can find that the irreversibility becomes significant when the maximum field is greater than 10 T. This phenomenon indicates that an energy dissipative process undergoes during the magnetization or demagnetization process. The degree of the energy dissipation, i.e., the amount of the heat production during pulsed fields, can be measured by the temperature difference (ΔT_{irr}) between the initial and the final temperatures. As seen in Fig. 6, ΔT_{irr} increases with increasing the maximum magnetic field (B_M) of the field pulse. The B_M dependence of ΔT_{irr} is plotted in Fig. 7. The ΔT_{irr} increases rapidly at B_M of around 10 T, which indicates that a spin gap closure in this field region closely relates to the energy dissipative phenomenon.

One may come up with the idea that magnetostriction can be a cause of the energy dissipation [21]. If magnetostriction occurs in a ms pulsed magnetic field, the long 1/4 plateau observed from 42 to 67 T is a feature of magnetization reflecting the lattice deformed by a magnetic field. More interestingly, the magnetostriction can depend on the field sweep rate of the magnetic field and a μs pulsed field can be too fast to induce the magnetostriction. Under the circumstance, reflecting the original lattice of spins, the 1/4 plateau region can shorten and the 1/3 plateau newly appears around 50 T.

To validate whether considerable magnetostriction occurs and if it occurs, to know how it behaves when the field sweep rate of magnetic field changes, magnetostriction measurements were conducted. Figures 8(a) to 8(d) show the results of the magnetostriction with different pulse duration of magnetic fields. The magnetostriction and magnetic fields are plotted as a function of time. The $\Delta L/L$ data are represented with dark-blue curves. We attribute part of the changes in the signal to the mechanical vibration because of two features. One is that it suddenly (in an indifferentiable way) occurs in contrast to that the magnetostriction is found to gradually happen. Another is that the effect of the vibration seems to occur at a specific time around 1 ms. The changes appear to be influenced by a

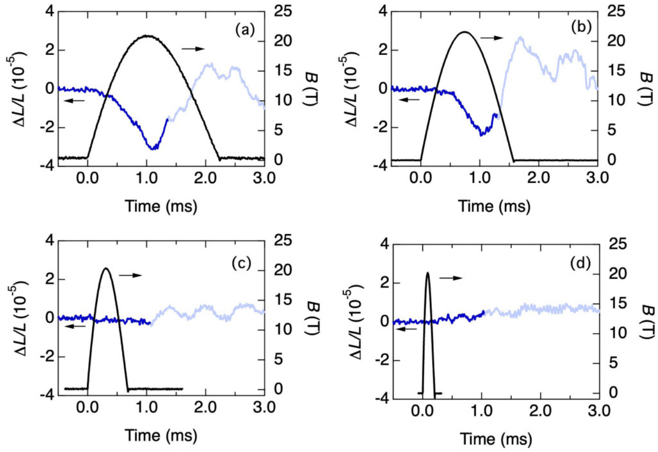

FIG. 8. The magnetostriction $\Delta L/L$ and the magnetic field are plotted as a function of time.

mechanical vibration due to the generation of a magnetic field are represented with light-blue curves.

In Fig. 8(a), magnetostriction($\Delta L/L$) is found to occur and reaches 3×10^{-5} at 21 T. The pulse duration of the magnetic field is 2.2 ms. When a pulse duration becomes shorter (1.6 ms) as shown in Fig. 8(b), the $\Delta L/L$ reaches only 2.2×10^{-5} with the same magnetic field strength of 21 T. Moreover, it is found that the response of the magnetostriction to the magnetic field is delayed. An even more striking phenomenon is that the magnetostriction seems to be absent when the pulse duration is shorter as shown in Figs. 8(c) and 8(d). The pulse durations are 0.7 and 0.2 ms for Figs. 8(c) and 8(d), respectively.

In Fig. 9, $\Delta L/L$ measured with a longer magnetic field pulse of up to 42 T is plotted as a function of magnetic field along with the results shown in Figs. 8(a) to 8(d). The duration time of the 42 T pulse is 36 ms. The rather large fluctuation of the data for 42 T is due to the small intensity of the light signal of the FBG that is caused by using a shorter length FBG at the measurement.

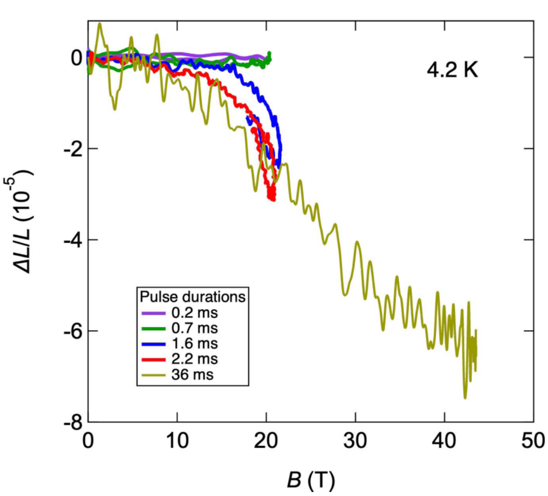

FIG. 9. $\Delta L/L$ measured with a longer magnetic field pulse of up to 42 T is plotted as a function of magnetic field along with the results shown in Figs. 8(a) to 8(d).

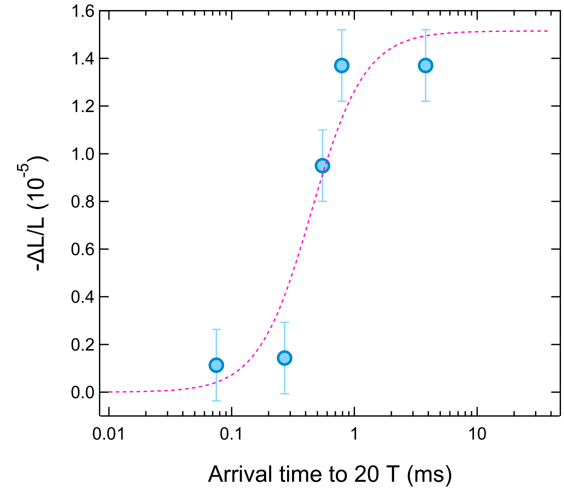

FIG. 10. $-\Delta L/L$ at 20 T is plotted as a function of a time required to reach 20 T. The dashed curve is a guide for eyes.

A steep increase of the negative magnetostriction is seen at around 20 T for the measurement of a 1.6-ms pulse and for that of 2.2-ms pulse. Also, the magnetostriction seems to behave with hysteresis in the field-descending process. This behavior is qualitatively understood in terms of the slow response of the magnetostriction. Because the time derivative of the magnetic field dB/dt becomes small near the top of the magnetic field pulse, it is expected that the magnetostriction can respond to the field change when the field reaches close to the top of the pulse. A steep change thus occurs near the top of the field pulse, and delayed response results in the hysteresis-like behavior.

In Fig. 10, $-\Delta L/L$ at 20 T is plotted as a function of a time required to reach 20 T for different magnetic field pulses. No magnetostriction is observed when the time is approximately shorter than 0.4 ms. Here, we can conclude that magnetostriction takes place under the condition that a field sweep rate is smaller than roughly 20 T/0.4 ms = 50 T /ms. The field sweep rate for the nondestructive 69 T pulse that was used for the measurement of the magnetization curve (green curve in Fig. 1) is evaluated to be around 69 T/ 2 ms, which satisfies the condition required for inducing magnetostriction. On the other hand, in the single-turn coil experiment, the field sweep rate is as large as 5.0×10^4 T/ ms, this is three orders of magnitude larger than the criteria and no magnetostriction is expected to occur.

It is not unreasonable to expect that the exchange interaction is modified by the magnetostriction and the magnetization curve should be different from that of the original lattice. Here we can regard the magnetization curve in the single-turn coil experiment (a red curve in Fig. 1) as the original magnetization curve, and the magnetization curve measured with a nondestructive magnet (the green curve in Fig. 1) shows a magnetization process influenced by the magnetostriction. To make a semi-quantitative discussion about the effect of magnetostriction on the magnetization curve, we calculated the magnetization curve using the density-matrix renormalization group (DMRG) method for $N = 60$ sites. For this calculation, we used the ALPS code library [22,23]. The

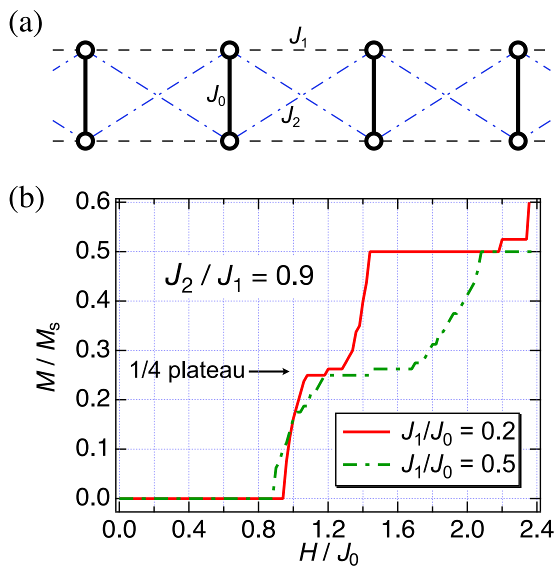

FIG. 11. (a) $S = 1$ ladder model with a diagonal interaction J_2. (b) Calculated results of the magnetization curve when $J_2/J_1 = 0.9$. Red solid line and green dot-dashed line denote the results for $J_1/J_0 = 0.2$ and $J_1/J_0 = 0.5$, respectively.

two major differences between the red and the green curves in Fig. 1 are the presence of the 1/3 plateau only in the red curve and the difference in the length of the 1/4 plateau between the two. Because the mechanism of the 1/3 plateau is still unclear and it is beyond the scope of the present work, we limit our discussion to the relationship between magnetostriction and the length of the 1/4 plateau. As an example of a spin-lattice model for an $S = 1$ spin ladder system which exhibits a 1/4 plateau, a model with a diagonal interaction J_2 in addition to J_0 (rung) and J_1 (leg) shown in Fig. 11(a) was proposed by Okamoto *et al.* [24]. Since the 1/4 plateau appears when J_1 and J_2 are similar magnitudes in this model, we fixed the ratio of J_1 and J_2 to $J_2/J_1 = 0.9$ in this calculation. From the magnetostriction measurement, it was found that the lattice contraction occurs along the leg direction. Therefore, we simulated a magnetization curve with increasing the magnitudes of J_1 and J_2 relative to J_0. The red solid line and the green dot-dashed line in Fig. 11(b) are the magnetization curves for $J_1/J_0 = 0.2$ and 0.5, respectively. It is found that an increase in J_1/J_0 from 0.2 to 0.5 stabilizes the 1/4 plateau; the field region of the 1/4 plateau becomes three times longer. This behavior reproduces the difference between the red and green curves in Fig. 1. Moreover, it should be noted that the increase in J_1/J_2 slightly shifts the field at which the 1/4 plateau starts to the high field side. This finding is also in good agreement with the experimental results. The present result of the simulation indicates that the increase in J_1 and J_2 due to the contraction along the leg direction can stabilize the 1/4 plateau and explain the dependence of the magnetization curve on the magnetic-field sweep rate. Here, it should be mentioned again that the theoretical model shown in the present work can only explain the behavior of the 1/4 plateau. It suggests that further theoretical investigation is necessary to elucidate details of the spin-lattice decoupling in BIP-TENO.

V. CONCLUSION

The $S = 1$ two-leg spin ladder compound BIP-TENO was studied with an ultrahigh magnetic field of up to 150 T. Magnetostriction is found to play an important role in stabilizing the 1/4 plateau when a magnetic field is swept in a millisecond timescale or longer. A high-speed magnetization process in a microsecond timescale can realize the adiabatic condition, i.e., a crystal lattice is stationary, and only spins respond to magnetic fields, which results in a separation of the spin and lattice. The full magnetization process of BIP-TENO is proposed based on the experimental results using a microsecond ultrahigh magnetic field of up to 150 T. The 1/3 and 1/2 plateaus are observed in addition to the previously reported 1/4 plateau. Moreover, the 2/3 and 3/4 plateaus are strongly suggested to appear in higher fields over 100 T. The proposed full magnetization process corresponds to the magnetization process of the original lattice of spins that are free from magnetostriction. It would be an intriguing question whether the nontrivial 1/3 plateau appears only as the metastable state or not. A nondestructive 100-T magnet that has a millisecond pulse duration can solve this issue. Further theoretical study on the mechanism of the relatively slow magnetostriction would be required to understand the details of the spin-lattice separation discovered in the present work.

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