HAL
open science

Giant volume magnetostriction in La0.85Ce0.15Fe12B6

L. V. B. Diop, Mehdi Amara, O. Isnard

To cite this version:

L. V. B. Diop, Mehdi Amara, O. Isnard. Giant volume magnetostriction in La0.85Ce0.15Fe12B6. Applied Physics Letters, 2021, 118 (26), pp.262409. 10.1063/5.0056511 . hal-03325781

HAL Id: hal-03325781

https://hal.science/hal-03325781v1

Submitted on 25 Aug 2021

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

Giant volume magnetostriction in $La_{0.85}Ce_{0.15}Fe_{12}B_6$

L.V.B. Diop[1†], M. Amara[2] and O. Isnard[2]

[1]*Université de Lorraine, CNRS, IJL, F-54000 Nancy, France*

[2] *Institut NEEL, Université Grenoble Alpes, CNRS, 25 rue des martyrs, F-38042 Grenoble, France*

We report the discovery of giant magnetostriction and huge negative thermal expansion (NTE) in the itinerant-electron metamagnetic compound $La_{0.85}Ce_{0.15}Fe_{12}B_6$. This system presents multiple magnetic transformations, antiferromagnetic-ferromagnetic (AFM-FM) and ferromagnetic-paramagnetic (FM-PM), driven by changes in both external field and temperature. The magnetic-field-dependent magnetostriction exhibits irreversible abrupt jumps at $T \leq 5$ K, while it evolves smoothly with applied field above 5 K. A giant positive volume magnetostriction of $\Delta V/V$ (20 K, 6 T) = 0.80% is observed across the field-induced first-order AFM-FM metamagnetic transition. It is further revealed that the FM-PM transition is accompanied by a remarkably large NTE with a coefficient of linear thermal expansion $\alpha_L = -34\times10^{-6}$ K^{-1} over a temperature window of $\Delta T \sim 63$ K.

Itinerant electron metamagnetism (IEM) is a very specific phenomenon that develops in some 3d-4f intermetallics. It has potentially important functionalities like huge magnetostriction, colossal magnetoresistance, and giant magnetocaloric effects[1-4]. One fascinating example of IEM is the intermetallic compound $LaFe_{12}B_6$ which occupies a special place among rare-earth iron-rich alloys. The ternary system $LaFe_{12}B_6$ exhibits exotic magnetic features and intriguing physical properties. Unconventional and discontinuous avalanche-like (or staircase-like) metamagnetic phase transitions were recently discovered in $LaFe_{12}B_6$[5-8]. This unique and peculiar multistep magnetization process is characterized by steep magnetization jumps followed by plateaus. Neutron powder diffraction investigations revealed an unusual amplitude-modulated antiferromagnetic structure described by a propagation vector **k** = (¼, ¼, ¼) and remarkably weak Fe moment of about 0.43 μ_B in the ground state[5]. In addition, $LaFe_{12}B_6$ displays a particularly low Néel temperature T_N = 36 K, for an Fe-rich intermetallic phase, a multicritical point in the complex magnetic phase diagram[5], both inverse and normal magnetocaloric effects[9], colossal spontaneous magnetization steps occurring after a very long incubation time when both the applied magnetic field and temperature are kept constant[7], and large pressure effects[10]. These singular features not only motivated the development of

† leopold.diop@univ-lorraine.fr

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

experiments under extreme conditions and of new theoretical models[10-14], but also emphasized the potential interest of $LaFe_{12}B_6$ in future low-temperature energy technologies. The itinerant-electron metamagnet $LaFe_{12}B_6$ realizes a rare playground for materials physics owing to the extreme sensitivity of its physical properties to reasonably weak external hydrostatic pressure[10] and chemical substitution[8] that yields the effect of "chemical pressure". Within the $RT_{12}B_6$ family (where R is a rare-earth atom and T stands for a 3d transition metal element Co or Fe), $LaFe_{12}B_6$ is the sole stable Fe-based compound of the 1:12:6 ternary system[13-15], whereas the RCo$_{12}$B$_6$ phases are stable along the whole rare-earth series. $LaFe_{12}B_6$ intermetallic is unique among the $RT_{12}B_6$ series of compounds in having an antiferromagnetic ground state with an ordering temperature much smaller than the Curie temperature of the Co-based RCo$_{12}$B$_6$ ferro- (R =Y, La-Sm) or ferri- (R = Gd-Tm) magnets (T_C =134 - 162 K)[15]. It is worth to notice that the Néel temperature of $LaFe_{12}B_6$ is an order of magnitude smaller when compared to the magnetic transition temperature of any rare-earth iron-rich binary alloy. Interestingly, extraordinary electronic transport and magnetotransport properties have been most recently reported in RCo$_{12}$B$_6$ intermetallics with R = Y, Gd and Ho[16].

In a previous magnetic study of the pseudo-ternary (La,Ce)$Fe_{12}B_6$ system, we have demonstrated that both paramagnetic (PM) and antiferromagnetic (AFM) states get converted into the ferromagnetic (FM) state via a magnetic-field-induced metamagnetic transition[8]. In this letter, we report on the existence of giant magnetoelastic effects across the magnetic transitions in $La_{0.85}Ce_{0.15}Fe_{12}B_6$ as investigated by means of linear thermal expansion and magnetostriction measurements.

The specimen employed in the present work was taken from the same batch of the sample used in our earlier magnetization investigation. Details on the preparation and subsequent characterization of the polycrystalline alloy of composition $La_{0.85}Ce_{0.15}Fe_{12}B_6$ can be found in Ref. 8. Linear thermal expansion (LTE) and magnetostriction experiments were performed using a high sensitivity capacitance dilatometer in a superconducting magnet producing a maximum magnetic field of 6 T. In terms of relative length variation, the sensitivity is better than 10^{-7}. In this experimental setup, the capacitance cell can be rotated around the vertical axis of the cryostat, while the external field is applied along a fixed horizontal direction. Zero-field cooled (ZFC) and field cooled (FC) measuring protocols were used for LTE experiments under applied magnetic fields. Isothermal magnetostriction curves were recorded for both directions, parallel ($\Delta L/L_{//}$) and perpendicular ($\Delta L/L_{\perp}$) to the applied magnetic field. From these magnetostriction isotherms, the anisotropic magnetostriction ($\Delta L/L_{//}$ - $\Delta L/L_{\perp}$) and

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

the volume magnetostriction ($\Delta V/V = \Delta L/L_{//} + 2\Delta L/L_{\perp}$) were assessed.

The LTE ($\Delta L/L$) curves, recorded between 2.5 and 150 K under various magnetic fields, are presented in Figs. 1–2. In zero magnetic field (Fig. 1) both $\Delta L/L$ functions, heating and cooling, of $La_{0.85}Ce_{0.15}Fe_{12}B_6$ are identical. The spontaneous ($\mu_0 H = 0$ T) LTE plot displays an anomaly at ≈ 34 K; this value is in excellent agreement with the Néel temperature obtained from the temperature dependence of the magnetization. In the absence of applied magnetic field, $\Delta L/L$ decreases upon cooling (positive thermal expansion, PTE) at temperatures above T_N, by contrast, below T_N the material expands as the temperature is lowered (negative thermal expansion, NTE). No discontinuity is observed at the Néel temperature, reflecting a spontaneous magnetostriction consistent with the second-order AFM-PM phase transformation.

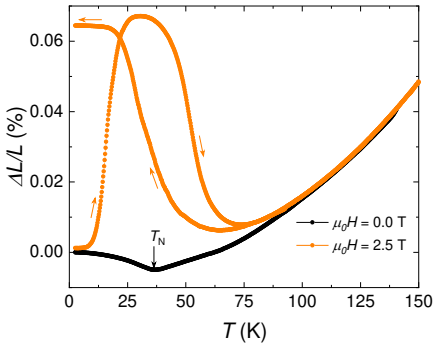


FIG. 1. Linear thermal expansion as a function of temperature recorded for $La_{0.85}Ce_{0.15}Fe_{12}B_6$ compound in magnetic fields of 0 and 2.5 T.

The thermal evolution of $\Delta L/L$ conducted in 2.5 and 3 T differs considerably from that seen in zero field. Upon heating from 2.5 K, $\Delta L/L$ exhibits a spectacular increase at the onset of the ferromagnetic ordering followed by a large drop and before increasing again at high temperature; leading to a bell-like curve. This peculiar length variation correlates with the presence of both low-temperature AFM-FM and high-temperature FM-PM transitions. The ZFC system shows a crossover from positive to negative thermal expansion because of the successive AFM-FM and FM-PM magnetic phase transformations. During the cooling process in 2.5 and 3 T, the lattice endures an expansion and then $\Delta L/L$ saturates at low temperatures. One observes a pronounced divergence between ZFC and FC modes. The temperature hysteresis accompanying the FM-PM transformation – Fig 1 – is remarkably large, ≈ 20 K for $\mu_0 H = 2.5$ T, and highlights the first-order nature of the phase transition.

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

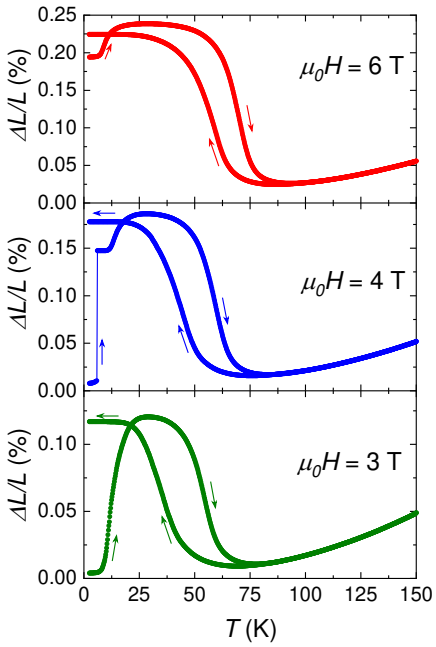


FIG. 2. Isofield linear thermal expansion of $La_{0.85}Ce_{0.15}Fe_{12}B_6$ compound as a function of temperature in applied magnetic fields of 3, 4 and 6 T.

Unexpectedly, the 4 T isofield LTE curve (Fig. 2) displays an abrupt jump at which the relative length varies suddenly from 0.01 to 0.15% when the temperature raises by only 0.2 K. This feature demonstrates that under a magnetic field of 4 T, the temperature change stimulates the order-order AFM→FM magnetic transformation at low temperatures. Furthermore, in a 6 T applied magnetic field, a prominent NTE phenomenon is detected over a temperature window of ΔT~63 K yielding an average linear thermal expansion coefficient $\alpha_L = (1/L)(\Delta L/\Delta T) = -34\times10^{-6}$ K^{-1}. This large NTE effect originates from the strong coupling between the magnetism and the crystal lattice. Our NTE coefficient compares well with the value obtained for the itinerant-electron system $LaFe_{10.5}CoSi_{1.5}$[17], $\alpha_L = -26\times10^{-6}$ K^{-1}. Note that the absolute value of α_L for the commercial NTE material ZrW_2O_8[18] ($\alpha_L = -9\times10^{-6}$ K^{-1}) is about 4 times smaller than that of $La_{0.85}Ce_{0.15}Fe_{12}B_6$ intermetallic compound.

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

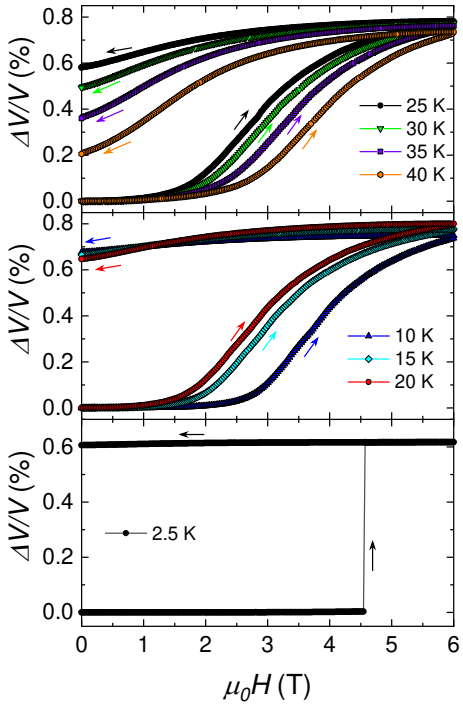


FIG. 3. Volume magnetostriction isotherms of $La_{0.85}Ce_{0.15}Fe_{12}B_6$ measured at the indicated temperatures ranging between 2.5 and 40 K.

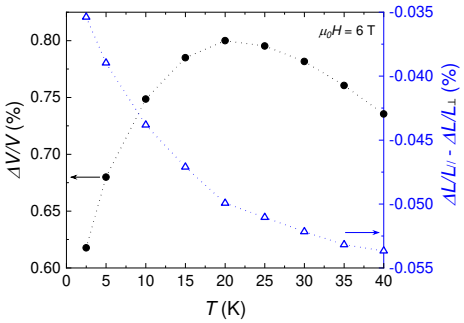


FIG. 4. Temperature dependence of the volume ($\Delta V/V = \Delta L/L_{//} + 2\Delta L/L_{\perp}$) and anisotropic (($\Delta L/L_{//}$ - $\Delta L/L_{\perp}$) magnetostriction at the maximum applied field of 6 T as recorded for $La_{0.85}Ce_{0.15}Fe_{12}B_6$.

To further elucidate the lattice response at the metamagnetic transition, longitudinal ($\Delta L/L_{//}$) and transverse ($\Delta L/L_{\perp}$) magnetostriction isotherms were measured at various fixed temperatures. The corresponding isothermal volume magnetostriction curves, $\Delta V/V$, are plotted

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

in Fig. 3. For each isotherm, the applied magnetic field was cycled between 0 and the maximum attainable value of 6 T. Each forced volume magnetostriction isotherm starts from the virgin state after cooling the sample in zero field from the paramagnetic region. During the first magnetic field increase at 2.5 K (bottom panel), $\Delta V/V$ presents an ultrasharp stepwise change of 0.61% at a certain critical field $\mu_0 H_{cr}$ = 4.5 T. This tremendous relative volume variation is caused by the magnetic-field-induced AFM-FM transition, with the AFM ground state having a lower volume when compared with that of the field-driven FM state. The subsequent reverse leg shows no jump and $\Delta V/V$ keeps a constant value down to the zero magnetic field point. The compound does not recover the initial volume value and stays in the final deformation state after reducing the applied magnetic field to zero (corresponding to the induced-FM state). This experimental observation reveals that the AFM-FM transformation is totally irreversible at very low temperature. To restore the original AFM ground state (virgin state), and thus, the initial volume value, the system should be heated above the ferromagnetic ordering temperature and subsequently cooled in zero magnetic field.

At slightly higher temperatures for instance 10 K and above (Fig. 3), $\Delta V/V$ displays gradual changes across both AFM-FM and PM-FM magnetic phase transformations unlike the discontinuous process observed at 2.5 K. The volume magnetostriction isotherms exhibit huge hysteresis against the magnetic field scan, which is one of the classical signatures of a first-order transition. This hysteretic character and the irreversible/reversible behavior depend on the temperature interval. Two distinct representative temperature ranges are illustrated in Fig. 3: between 10 and 20 K (middle panel) and $T \geq 25$ K (top panel). In the AFM phase, for example at 10 K, the relative volume change is very small at low magnetic fields but dramatically grows as the material undergoes a phase transition to the FM state giving rise to a giant positive magnetostriction effect. No transition is detected throughout the decreasing-field run and $\Delta V/V$ remains essentially constant. This is reflecting the irreversible nature of the transition at temperatures lower than 25 K. Above 25 K, the curve of the downward-field process deviates from the nearly pure FM behavior and a metamagnetic-like transition takes place at lower field, producing a clear magnetic hysteresis between both field scans. Above 25 K, the magnetic-field-driven AFM-FM and PM-FM transformations are partially reversible; a proportion of the alloy recovers the initial AFM or PM state after removal of the applied field. In other words, both reversible and irreversible transitions coexist in this temperature interval, and the concentration of the sample, which endures the irreversible transformation, diminishes upon heating. At temperatures exceeding 25 K, the transition field determined for the ascending-field

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

branch increases upon heating while it shows the opposite trend at lower temperatures. Below 25 K, the critical field of the first-order AFM-FM transformation rises with decreasing the temperature owing to the strengthening of the negative exchange interactions and the diminution of the thermal fluctuations of the magnetic moments and elasticity of the lattice in the AFM phase. This yields the increase of both the free energy difference between the two magnetically ordered AFM and FM phases, and the critical field needed to accomplish the magnetic transformation from one state to another.

The temperature dependence of the anisotropic and volume magnetostriction at the maximum attainable magnetic field of 6 T is represented in Fig. 4. In $La_{0.85}Ce_{0.15}Fe_{12}B_6$ a highly anisotropic effect is found, i.e., the value of the transverse magnetostriction is larger than that of the longitudinal one ($\Delta L/L_{\perp} > \Delta L/L_{//}$). The anisotropic magnetostriction is as large as -0.05% at 20 K and 6 T. Yet the isothermal magnetostriction curves are not saturated even under 6 T. For example, at 2.5 K the applied magnetic field needs to increase up to 9.3 T for the compound to reach the fully FM polarized state[8], however, this field strength was not reachable on our magnetostriction set-up. The forced volume magnetostriction associated with the field-induced first-order AFM-FM transition amounts to $\Delta V/V$ (20 K, 6 T) = 0.80%. This later value is reasonably in accord with the relative volume variation estimated from LTE data of Fig. 2 (top panel), $\Delta V/V = 3\Delta L/L = 0.64\%$, particularly considering the anisotropic character. The magnitude of the volume change, $\Delta V/V$ (20 K, 6 T) = 0.80%, in the itinerant-electron compound $La_{0.85}Ce_{0.15}Fe_{12}B_6$ is comparable to the isotropic forced volume magnetostriction reported for iron-rich alloys such as $La(Fe_xAl_{1-x})_{13}$ and $La(Fe_xSi_{1-x})_{13}$. Indeed, a relative volume variation of about 1% was found in $La(Fe_{0.87}Al_{0.13})_{13}$ intermetallic[19]. Volume magnetostriction of 0.9% was obtained for the pseudo-binary $La(Fe_{0.86}Si_{0.14})_{13}$ alloy[20].

The strong magnetic-field-driven irreversibility of the volume magnetostriction curves is an outstanding feature and consistent with the irreversible behavior seen in isothermal magnetization. The sharp discontinuities in the linear thermal expansion and magnetostriction isotherms strikingly resemble to those observed in the magnetic data of Ref. 8 and proves the strong correlation between elastic properties and magnetism in $La_{0.85}Ce_{0.15}Fe_{12}B_6$. Our LTE and magnetostriction results clearly demonstrates that the magnetic transitions are accompanied by huge magnetovolume effects. At fixed temperatures, the applied magnetic field triggers a transformation from a low-volume low-magnetization to a high-volume high-magnetization phase.

In conclusion, the itinerant-electron metamagnetic system $La_{0.85}Ce_{0.15}Fe_{12}B_6$ exhibits

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

many salient features in the LTE and magnetostriction data including a crossover from positive thermal expansion to negative one, a remarkably large thermal hysteresis, a strong magnetic-field-driven irreversibility, a huge magnetic field hysteresis and ultrasharp steps. A giant volume magnetostriction $\Delta V/V = 0.80\%$ has been found in $La_{0.85}Ce_{0.15}Fe_{12}B_6$ making this alloy attractive as cryogenic magnetostrictive materials. However, the potential technological applications of this compound in cryogenic magnetostrictive actuator are limited by the relatively large critical fields and the huge magnetic hysteresis at low temperature. In addition, a large negative thermal expansion effect with a linear thermal expansion coefficient $\alpha_L = -34\times10^{-6}$ K^{-1} was discovered. Our experimental findings shed light on the strong coupling between lattice and magnetic degrees of freedom in $La_{0.85}Ce_{0.15}Fe_{12}B_6$.

DATA AVAILABILITY

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

REFERENCES

[1]D.P. Kozlenko, E. Burzo, P. Vlaic, S.E. Kichanov, A.V. Rutkauskas, and B.N. Savenko, Sci. Rep. **5**, 8620 (2015).

[2]A. Fujita, S. Fujieda, Y. Hasegawa, and K. Fukamichi, Phys. Rev. B **67**, 104416 (2003).

[3]H. Yamada, and T. Goto Phys. Rev. B **68**, 184417 (2003).

[4]N.H. Duc, D.T. Kim Anh, and P.E. Brommer, Physica B **319**, 1 (2002).

[5]L.V.B. Diop, O. Isnard, and J. Rodríguez-Carvajal, Phys. Rev. B **93**, 014440 (2016).

[6]S. Fujieda, K. Fukamichi, and S. Suzuki, J. Magn. Magn. Mater. **421**, 403 (2017).

[7]L.V.B. Diop, and O. Isnard, Appl. Phys. Lett. **108**, 132401 (2016).

[8]L.V.B. Diop, and O. Isnard, Phys. Rev. B **97**, 014436 (2018).

[9]L.V.B. Diop, and O. Isnard, J. Appl. Phys. **119**, 213904 (2016).

[10]L.V.B. Diop, O. Isnard, Z. Arnold, J.P. Itié, J. Kastil, and J. Kamarad, Solid State Comm. **252**, 29 (2017).

[11]G.I. Miletic, and Z. Blazina, J. Magn. Magn. Mater. **323**, 2340 (2011).

[12]G.I. Miletic, and Z. Blazina, J. Alloys Compd. **430**, 9 (2007).

[13]M. Rosenberg, T. Sinnemann, M. Mittag, and K.H.J. Buschow, J. Alloys Compd. **182**, 145 (1992).

[14]Q.A. Li, C.H. de Groot, F.R. de Boer, and K.H.J. Buschow, J. Alloys Compd. **256**, 82 (1997).

[15]M. Mittag, M. Rosenberg, and K.H.J. Buschow, J. Magn. Magn. Mater. **82**, 109 (1989).

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

[16]F. Mesquita, S.G. Magalhaes, P. Pureur, L.V.B. Diop, and O. Isnard, Phys. Rev. B **101**, 224414 (2020).

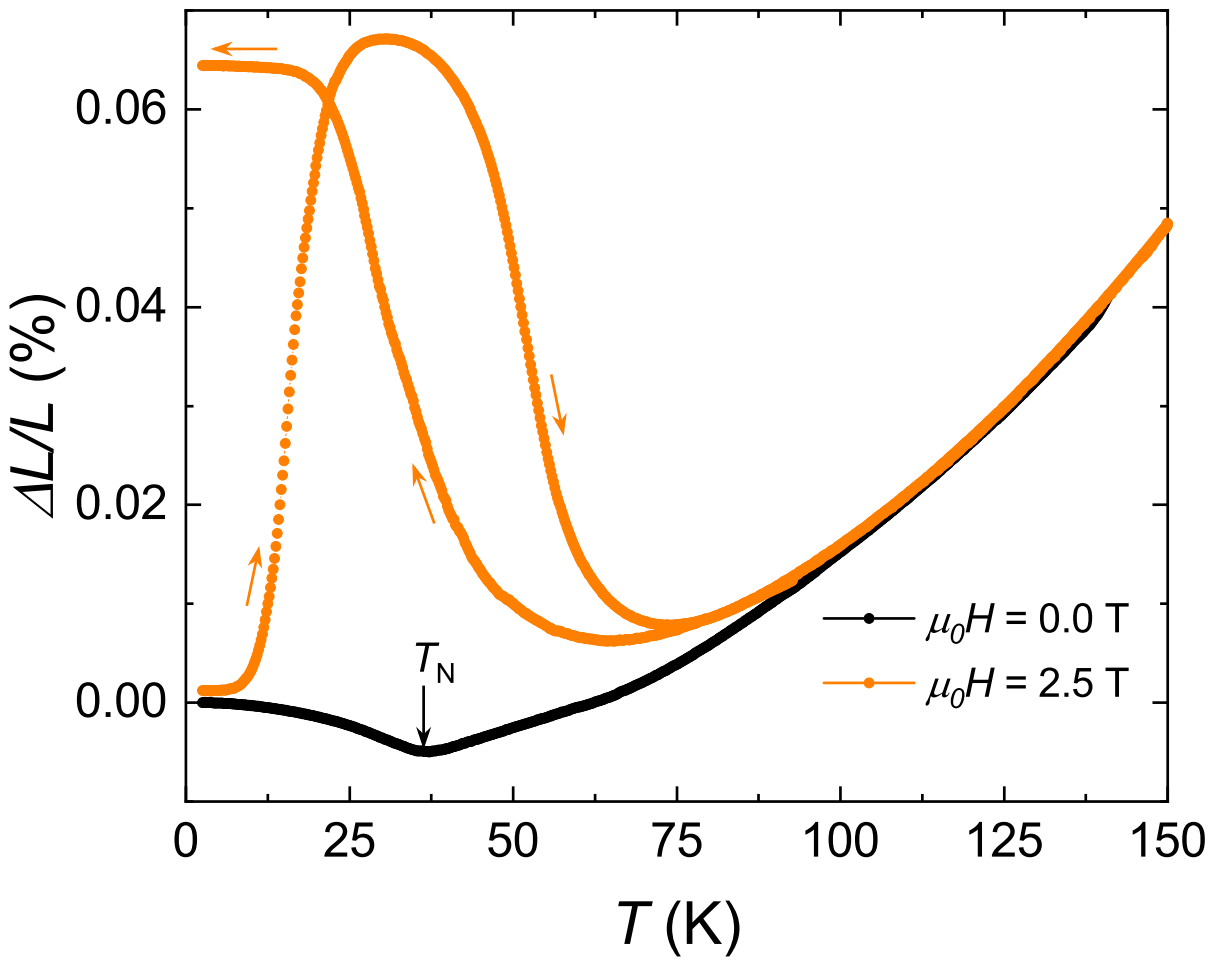
[17]R. Huang, Y. Liu, W. Fan, J. Tan, F. Xiao, L. Qian, and L. Li, J. Am. Chem. Soc. **135**, 11469 (2013).

[18]A.W. Sleight, Inorg. Chem. **37**, 2854 (1998).

[19]K. Irisawa, A. Fujita, K. Fukamichi, M. Yamada, H. Mitamura, T. Goto, and K. Koyama, Phys. Rev. B **70**, 214405 (2004).

[20]A. Fujita, Y. Akamatsu, and K. Fukamichi, J. Appl. Phys. **85**, 4756 (1999).

Applied Physics Letters ACCEPTED MANUSCRIPT
This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.
PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511
AIP Publishing

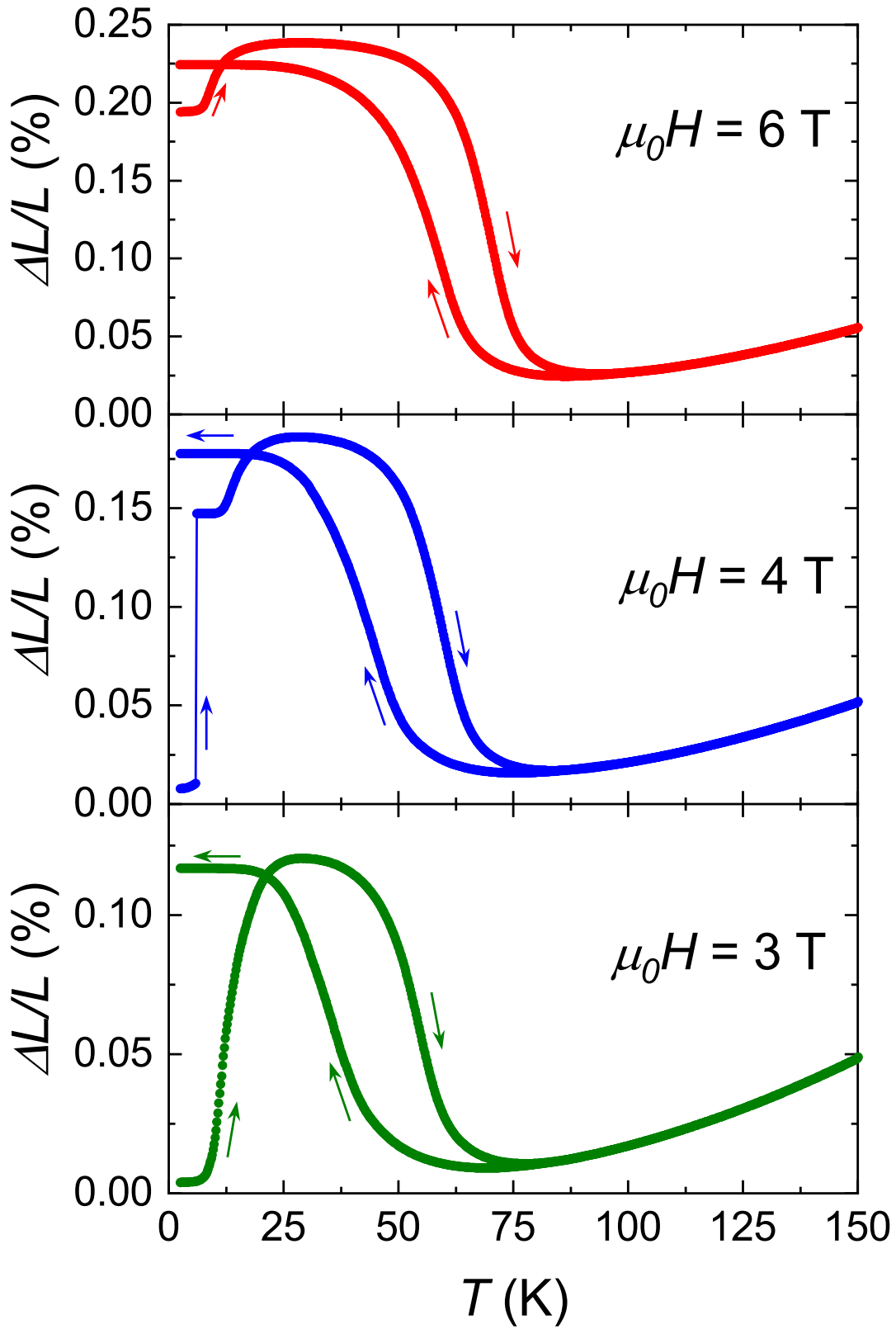


Applied Physics Letters ACCEPTED MANUSCRIPT

This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511

AIP Publishing

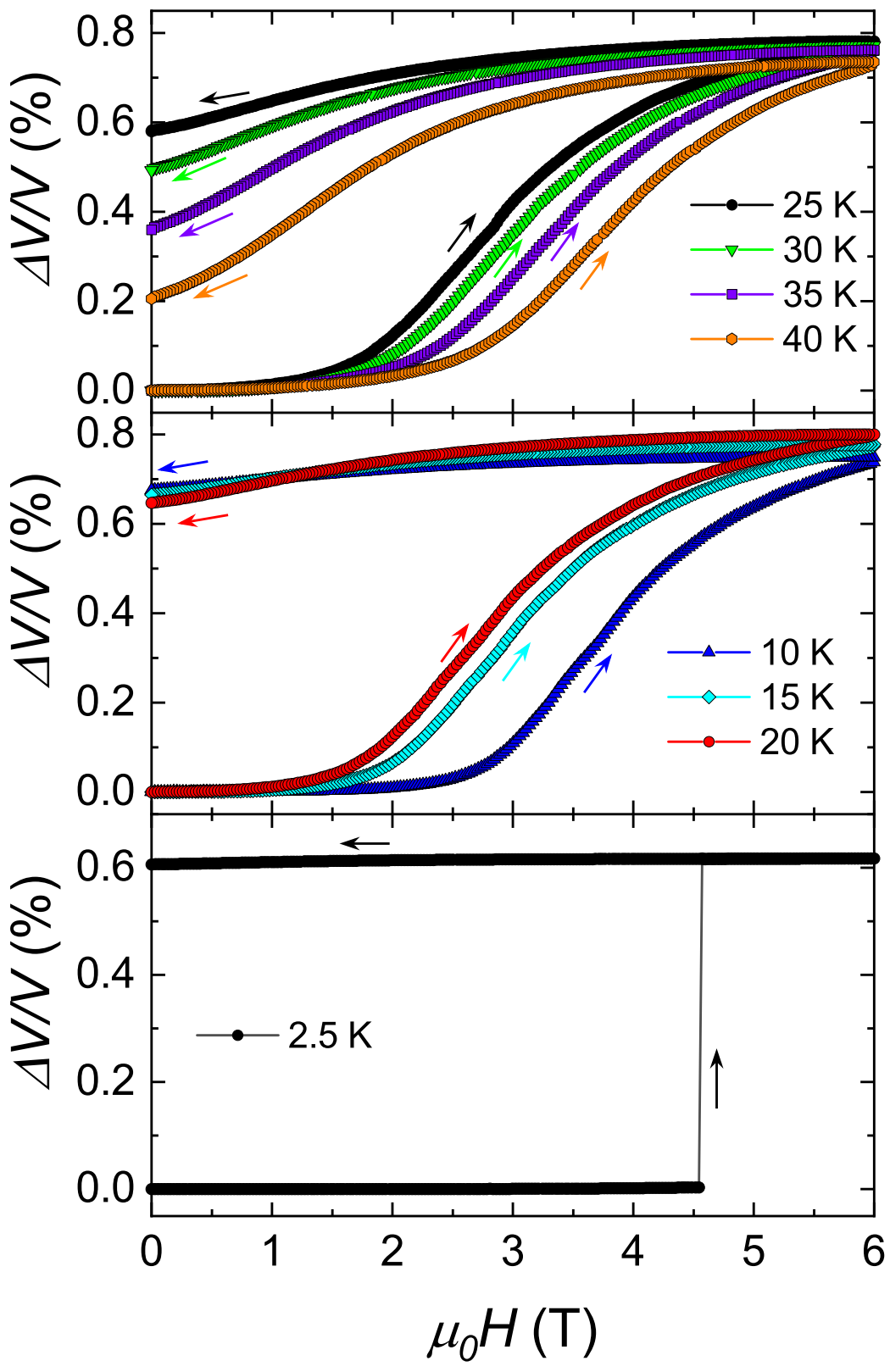


Applied Physics Letters ACCEPTED MANUSCRIPT

This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511

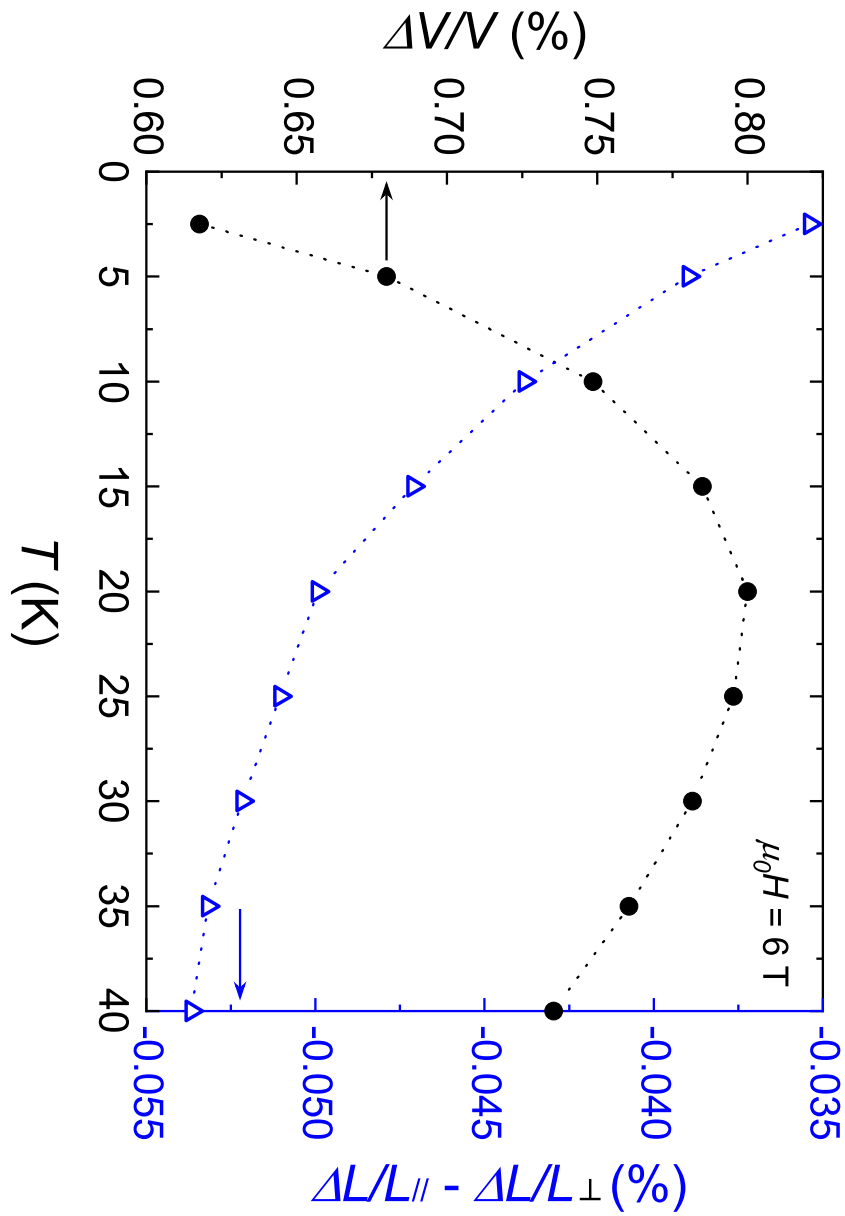
AIP Publishing



AIP Publishing

Applied Physics Letters

ACCEPTED MANUSCRIPT

This is the author's peer reviewed, accepted manuscript. However, the online version of record will be different from this version once it has been copyedited and typeset.

PLEASE CITE THIS ARTICLE AS DOI: 10.1063/5.0056511