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Origin of the 30 T transition in CeRhIn$_5$ in tilted magnetic fields

S. Mishra,$^1$ D. Gorbunov,$^2$ D. J. Campbell,$^3$ D. LeBoeuf,$^1$ J. Hornung,$^2$ J. Klotz,$^2$ S. Zherlitsyn,$^2$ H. Harima,$^4$ J. Wosnitza,$^{2,3}$ D. Aoki,$^5$ A. McCollam,$^6$ and I. Sheikin$^{1,7}$

$^1$Laboratoire National des Champs Magnétiques Intenses (LNCMI-EMFL), CNRS, UGA, 38042 Grenoble, France
$^2$Hochfeld-Magnetlabor Dresden (HLD-EMFL) and Würzburg-Dresden Cluster of Excellence ct.qmat, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany
$^3$Institut für Festkörper- und Materialphysik, TU Dresden, 01062 Dresden, Germany
$^4$Graduate School of Science, Kobe University, Kobe 657-8501, Japan
$^5$Institute for Materials Research, Tohoku University, Oarai, Ibaraki, 311-1313, Japan
$^6$High Field Magnet Laboratory (HFML-EMFL), Radboud University, 6525 ED Nijmegen, The Netherlands

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We present a comprehensive ultrasound study of the prototypical heavy-fermion material CeRhIn$_5$, examining the origin of the enigmatic 30 T transition. For a field applied at $2^\circ$ from the $c$ axis, we observed two sharp anomalies in the sound velocity, at $B_m \approx 20$ T and $B^* \approx 30$ T, in all the symmetry-breaking ultrasound modes at low temperatures. The lower-field anomaly corresponds to the well-known first-order metamagnetic incommensurate-to-commensurate transition. The higher-field anomaly takes place at 30 T, where an electronic-nematic transition was previously suggested to occur. Both anomalies, observed only within the antiferromagnetic state, are of similar shape, but the corresponding changes of the ultrasound velocity have opposite signs. Based on our experimental results, we suggest that a field-induced magnetic transition from a commensurate to another incommensurate antiferromagnetic state occurs at $B^*$. With further increasing the field angle from the $c$ axis, the anomaly at $B^*$ slowly shifts to higher fields, broadens, and becomes smaller in magnitude. Traced up to $30^\circ$ from the $c$ axis, it is no longer observed at $40^\circ$ below 36 T.

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I. INTRODUCTION

Unusual magnetic-field-induced transitions have recently become a matter of considerable interest in strongly correlated electron systems. In some heavy-fermion compounds, intermetallic materials based on rare earths or actinides, such transitions are rather exotic in nature. Among them are field-induced quantum critical points [1,2], field-induced or reinforced unconventional superconductivity [3–5], and Lifshitz transitions [6–8]. Understanding the mechanisms and the origins of such transitions is of primary importance in modern condensed matter.

CeRhIn$_5$, although discovered barely two decades ago [9], is one of the best studied heavy-fermion materials. This compound crystallizes into a tetragonal HoCoGa$_5$ type crystal structure (space group 123, $P4/mmm$) shown in Fig. 1(a). CeRhIn$_5$ undergoes an antiferromagnetic (AFM) transition at $T_N \approx 3.8$ K. The zero-field magnetic structure, AFM1, shown in Fig. 1(b), is incommensurate (IC) with propagation vector $\mathbf{Q}_{IC} = (1/2, 1/2, 0.297)$ [10,11]. The Ce magnetic moments are antiferromagnetically aligned within the basal plane and spiral transversally along the $c$ axis.

At low temperatures, when a magnetic field $B$ is applied in the basal plane, a first-order transition occurs at $B_m \approx 2$ T. The transition corresponds to a change of magnetic structure from IC AFM1 to commensurate (C) AFM3 with the propagation vector $\mathbf{Q}_C = (1/2, 1/2, 1/4)$ [12,13]. It is a collinear square configuration with “up-up-down-down” alignment. The magnetic moments are antiferromagnetically aligned in the basal plane along the direction perpendicular to the magnetic field, as shown in Fig. 1(c). When the magnetic field is tilted away from the basal plane, $B_m$ shifts to higher fields and follows the $1/\cos \alpha$ dependence ($\alpha$ is the field angle from the basal plane) up to about $75^\circ$–$80^\circ$. At higher angles, however, $B_m$ deviates from a $1/\cos \alpha$ dependence towards lower fields. The transition was traced up to $\theta \approx 2^\circ$ from the $c$ axis ($\alpha \approx 88^\circ$), where it occurs at $B_m \approx 20$ T [14].

Recent results obtained in high magnetic fields applied along or close to the $c$ axis suggest a remarkably novel behavior in CeRhIn$_5$. The AFM order is suppressed at $B \approx 50$ T giving rise to a field-induced quantum critical point [15,16]. Furthermore, Moll et al. [17] reported an observation of a hysteretic jump in the in-plane resistivity of CeRhIn$_5$ microstructures fabricated by focused ion beam (FIB) at $B^* \approx 30$ T along or close to the $c$ axis. Later on, these experiments were advanced, allowing for simultaneous measurements of the in-plane resistivity along the $[100]$ and $[010]$, as well as along the $[110]$ and $[\bar{1}0\bar{1}]$ directions [18]. These measurements revealed a strong in-plane resistivity anisotropy within each of the two inequivalent symmetry channels $B_{lg}$ ([100], [010]) and $B_{gg}$ ([110], [1\bar{1}0]), which emerges above $B^* \approx 30$ T tilted from the $c$ axis towards the [100] and [110] directions, respectively. This electronic anisotropy was interpreted in terms of an electronic-nematic transition. To the best of our knowledge, these results, however, have not been reproduced on bulk samples so far.
For a long time, all the attempts to detect an anomaly at $B^*$ in various measurements on bulk single crystals remained ineffective. In particular, no noticeable anomaly was observed at $B^*$ in longitudinal magnetization [19] or reported in magnetic-torque data [17,18]. It is only very recently that a small magnetostriction anomaly was observed at $B^*$ in a magnetic field higher than 2 T applied along the [100] direction (C). Arrows indicate the orientation of the magnetic moments. Only Ce atoms are shown in (b) and (c) for clarity. (d) Schematic illustration of the symmetry strains, $\varepsilon_{ij}$, induced by different ultrasound modes for a tetragonal crystal structure. For each mode, the propagation ($k$) and polarization ($u$) directions are shown by arrows. The associated irreducible representations are shown in brackets.

For all modes except $C_{33}$, the orientation of the magnetic field was determined from the frequency of sound velocities at $36 \, T$ [24]. The latter was observed in the $C_T = (C_{11} - C_{12})/2$ mode only, which induces the symmetry strain $\varepsilon_{xx} - \varepsilon_{yy}$ associated with the irreducible representation $B_{1g}$. This strain breaks the $C_4$ symmetry [Fig. 1(d)]. Another strain, which breaks the $C_4$ symmetry, is $\varepsilon_{xy}$. This strain, induced in the $C_{6g}$ mode, is associated with the $B_{2g}$ symmetry channel [Fig. 1(d)]. If the anomaly at $B^*$ is attributed to the previously suggested electronic-nematic transition [18], it is surprising that no anomaly was observed in the $C_{6g}$ and $C_{11}$ modes. These inconsistencies motivated us to re-examine the elastic response of CeRhIn$_5$ in high magnetic fields.

In this paper, we report high-resolution ultrasound-velocity measurements in CeRhIn$_5$ in high pulsed and static magnetic fields tilted away from the $c$ axis. We observed clear field-induced anomalies at both $B_m$ and $B^*$ in all symmetry-breaking modes. This suggests that the transition at $B^*$ is bulk in origin contrary to what was previously suggested. Both anomalies are of similar shape, but of the opposite sign. Both of them exist within the AFM state only. Furthermore, when the magnetic field is tilted further away from the $c$ axis, the anomaly at $B^*$ becomes broader and smaller in magnitude. It was traced up to an angle tilted by $30^\circ$ away from the $c$ axis.

### II. EXPERIMENTAL DETAILS

High-quality single crystals of CeRhIn$_5$ used in this study were grown by the In-self-flux technique, details of which can be found elsewhere [25]. Ultrasound velocity measurements were performed in both pulsed (up to 60 T) and static (up to 36 T) magnetic fields. In pulsed fields, we used a $^3$He flow cryostat, while static-field measurements were performed in a $^3$He cryostat equipped with an in situ rotator. In both cases, the measurements were performed using a pulse-echo technique in transmission mode. A pair of piezoelectric transducers was glued to opposite well-polished surfaces of the sample in order to generate and detect acoustic waves. The ultrasound frequencies and the absolute values of sound velocities at 4.2 K for different acoustic modes are given in Table I.

For all modes except $C_{33}$, the orientation of the magnetic field with respect to the $c$ axis was determined from the position of the anomaly at $B_m$, whose angular dependence was established in the previous torque measurements [14]. For the $C_{33}$ mode, the orientation was determined from the frequencies of the magnetoacoustic quantum oscillations, which were compared to the previously established angular dependence.
III. RESULTS AND DISCUSSION

Figure 2 shows low-temperature relative ultrasound velocity, $\Delta v/v$, as a function of $B$ applied close to the $c$ axis for several ultrasound modes. Two distinct anomalies are observed at $B_m \approx 20$ T and $B^* \approx 30$ T in all symmetry-breaking modes, i.e., $C_{11}$, $C_{44}$, $C_{66}$, and $C_T$. Both anomalies are absent in the $C_{33}$ mode, which does not break any tetragonal symmetry.

The non-symmetry-breaking $(C_{11} + C_{12})/2$ mode related to the $\epsilon_{zx} + \epsilon_{zy}$ strain belonging to the $A_{1g}$ representation cannot be measured directly using the pulse-echo technique. The relative ultrasound velocity variation for this mode was, therefore, obtained from the $C_{11}$ and $C_T$ data. First, the absolute values of the zero-field elastic constants were calculated as $C_0 = \rho v^2$ using the ultrasonic velocities $v$ given in Table I and the density $\rho = 8.316 \text{ g/cm}^3$. Then, the field-dependent elastic constant variations were calculated as $\Delta C(B) = 2 \frac{\Delta \rho}{\rho} C_0$ using the measured field dependence of $\Delta v/v$ for both $C_{11}$ and $C_T$ modes, shown in Figs. 2(a) and 2(b), respectively. Since $(C_{11} + C_{12})/2 = C_{11} - C_T$ and $\Delta(C_{11} + C_{12})/2 = \Delta C_{11} - \Delta C_T$, the field dependence of $\Delta C/C$ for the $(C_{11} + C_{12})/2$ mode was obtained. Finally, the relative ultrasound velocity variation was calculated as $\Delta v/v = (\Delta C/C)/2$.

The resulting velocity variation for the $(C_{11} + C_{12})/2$ mode is shown in Fig. 3. Here, the line thickness represents the resulting error due to the uncertainties in the determination of the ultrasound velocities for the $C_{11}$ and $C_T$ modes. One can see that, similar to the $C_{33}$ mode, the $(C_{11} + C_{12})/2$ mode does not show any apparent anomalies within the experimental error.

As mentioned above, both anomalies in the symmetry-breaking modes have a similar shape, but the corresponding changes of $\Delta v/v$ are of opposite sign. In the $C_{11}$ and $C_T$ modes, the anomalies at $B_m$ and $B^*$ appear as a sharp decrease and increase of the ultrasound velocity, respectively. These anomalies are remarkably similar to those previously observed in other compounds, in which field-induced spin-reorientation transitions occur [26–30]. In the $C_{44}$ and $C_{66}$ modes, both characteristic fields result in similar but somewhat reduced features. Another anomaly, a minimum in $\Delta v/v$ versus $B$, is clearly visible at $B_p \approx 44$ T in the $C_{11}$ mode. This anomaly was also observed in the previous ultrasound study, where it was interpreted as a transition (or crossover) into the polarized paramagnetic state [24].
The anomaly at \( B_m \) corresponds to the well-known first-order metamagnetic transition, where the magnetic structure changes from the IC AFM1 to the C AFM3 phase, as schematically shown in Fig. 1. Since the transition is of the first order, it is not surprising that it manifests itself in most of the ultrasound modes.

Regarding the transition at \( B^* \), its observation in all symmetry-breaking modes suggests that the transition is probably also of the first order, in agreement with recent specific heat measurements [23]. As mentioned, the anomalies at \( B_m \) and \( B^* \) have similar shapes but opposite signs. It is, therefore, natural to conclude that both transitions are of the same origin, i.e., that the transition at \( B^* \) corresponds to another field-induced change of the magnetic structure, this time from the C AFM3 to an IC AFM4 phase. This conclusion is in line with previous NMR results, which unambiguously suggest an IC phase above \( B^* \) [21].

The above hypothesis regarding the origin of the transition at \( B^* \) is further supported by the measurements performed at different temperatures, as shown in Fig. 4 for the \( C_{11} \) (a), \( C_T = (C_{11} - C_{12})/2 \) (b), \( C_{44} \) (c), and \( C_{66} \) (d) acoustic modes. In all cases, the magnetic field was applied at \( \theta \approx 2^\circ \). The data for the \( C_T \) mode [Fig. 4(b)] were obtained in static fields. All the other curves were measured in pulsed magnetic fields.

The anomalies at both \( B_m \) and \( B^* \) remain sharp up to the highest temperature, at which they were observed. The anomaly at \( B_m \) is observed up to higher temperatures than its counterpart at \( B^* \). Both anomalies are almost temperature independent at low temperatures. At temperatures approaching the antiferromagnetic phase boundary (\( \sim 2.5 \) K for \( B_m \) and \( \sim 2.4 \) K for \( B^* \)), \( B_m \) and \( B^* \) shift to slightly higher and lower field, respectively.

Both anomalies from all the measurements are traced in the resulting phase diagram together with the AFM boundary from the previous specific-heat results [23] (see Fig. 5). In agreement with previous reports [18,20,24], both anomalies at \( B_m \) and \( B^* \) are observed within the AFM state only, suggesting that both transitions are related to the magnetic properties of CeRhIn5. This assumption is strongly supported by the presence of clear anomalies, both at \( B_m \) and \( B^* \), in magnetic torque divided by field, \( \tau/|B| \), shown in Fig. 6.

Next, we discuss the angular dependence of the transition fields \( B_m \) and \( B^* \). The latter was obtained from measurements of the \( C_T \) mode, where both anomalies are most pronounced. The field dependence of \( \Delta v/v \) for different angles \( \theta \) from [001] towards the [110] direction is shown in Fig. 7(a). At 2\(^\circ\), both anomalies manifest themselves as sharp steps [Fig. 7(b)]. With increasing angle, both features progressively become smaller. While the anomaly at \( B_m \) remains sharp, the one at \( B^* \) broadens with increasing angle, as evidenced by a progressive small increase of the FWHM of the first derivative shown in the inset of Fig. 7(d). This probably implies that the transition at \( B^* \) at small angles changes to a crossover at larger angles. This assumption is supported by recent specific-heat [23] and magnetostriction [20] measurements. While the specific heat, performed in magnetic fields applied parallel or very close to the \( c \) axis, suggests a phase transition, the magnetostriction, performed in fields tilted by \( \sim 20^\circ \), suggests a crossover at \( B^* \). The anomaly at \( B_m \) is easily traceable all the way up to 40\(^\circ\), while its counterpart at \( B^* \) is still present at 30\(^\circ\), but is no longer visible at 40\(^\circ\), where the curve is completely dominated by strong low-frequency quantum oscillations [Fig. 7(e)]. The angular dependence of the anomaly at \( B^* \) is strikingly different from that observed in resistivity measurements on FIB-fabricated microdevices [17,18], where the resistivity jump at \( B^* \) remained sharp all the way up to 60\(^\circ\), the highest angle to which it was traced. The size of the jump showed a nonmonotonic behavior with a maximum at about 20\(^\circ\) from the \( c \) axis. This difference between bulk samples and microfabricated devices is probably due to
FIG. 5. $B - T$ phase diagram of CeRhIn$_5$ obtained from the ultrasound-velocity anomalies observed in all modes. Triangles correspond to the AFM to paramagnetic (PM) transition from Ref. [23]. AFM1 and, presumably, AFM4 are IC phases with different propagation vectors. AFM3 is the C phase discussed in the text. PPM stands for the polarized paramagnetic phase. The solid line is a fit of $T_N(B) = T_N(0)[1 - (B/B_c)^2]$ to the Néel temperature, with $B_c \approx 52$ T. Dashed lines are guides for the eye.

uniaxial stresses or strains inevitably present in FIB-fabricated devices [31]. CeRhIn$_5$ seems to be very sensitive to uniaxial strains, especially to orthorhombic strains, as evidenced by a particularly large size of the anomalies in the CT mode. Furthermore, previously reported NQR measurements on bulk and powder samples of CeRhIn$_5$ suggest that even small strains (or stresses) change the zero-field magnetic structure from IC to C [32]. Additional measurements under uniaxial stress (or strain) at high magnetic fields on bulk samples are required to elucidate the role of the uniaxial stress in previous transport measurements on CeRhIn$_5$ microstructures [17,18].

The resulting angular dependence of both $B_m$ and $B^*$ is shown in Fig. 7(d). In agreement with previous results [14], $B_m$ is strongly angle dependent. It is well fit by $1/\cos(90^\circ - \theta)$ down to about 10$^\circ$ from the c axis but deviates at lower angles. $B^*$, on the other hand, is only weakly angle dependent. It increases approximately linearly all the way from 2$^\circ$ to about 30$^\circ$, above which it either disappears or shifts to beyond 36 T. The remarkable difference between the two angular dependencies is probably due to different magnetic moment arrangements in the vicinity of the two transitions. While the moments are aligned in the basal plane in the vicinity of $B_m$, they are probably tilted towards the c axis close to $B^*$.

Finally, we comment on the magnetoacoustic quantum oscillations. In agreement with the results of the previous high-field ultrasound study of CeRhIn$_5$ [24], we observed strong low-frequency magnetoacoustic quantum oscillations in the CT mode between $B_m$ and $B^*$, as shown in the inset of Fig. 8. In this mode, these oscillations with a frequency

FIG. 6. Magnetic torque divided by field, $\tau/B$, vs $B$ applied at $\theta = 2^\circ$ from [001] towards the [110] direction at $T = 30$ mK.

FIG. 7. (a) $\Delta \nu/\nu$ for the CT mode as a function of $B$ applied at different angles, $\theta$, from the c axis at $T = 1.25$ K. Curves are vertically shifted for clarity. (b) and (c) show the curves at the two smallest and largest angles of our measurements, respectively. (d) The angle dependence of the transition fields $B_m$ and $B^*$ determined from the minima (maxima) of the first derivative, $d(\Delta \nu/\nu)/dB$, an example of which is shown in the inset. The solid line is a $1/\cos(90^\circ - \theta)$ fit to the data above 10$^\circ$. The dashed line is a guide for the eye.
FIG. 8. Fast Fourier transform (FFT) spectra of the magnetoacoustic quantum oscillations in the \( C_T \) mode (shown in the inset) both below and above \( B^* \). A nonoscillating background was subtracted prior to performing the FFTs. The FFTs were performed over equidistant \( 1/B \) ranges indicated by rectangles in the inset.

denoted \( A (F_A \simeq 0.6 \, \text{kT}) \) disappear above \( B^* \), as shown in Fig. 8, also in agreement with the previous study. This experimental result was interpreted as evidence of a possible Fermi-surface reconstruction at \( B^* \) in the previous work [24]. Similarly, additional oscillatory frequencies emerging above \( B^* \) were observed in the dHvA effect [15,22] and magnetostriction [20]. The appearance of these additional frequencies was discussed in terms of the Fermi-surface reconstruction at \( B^* \) due to \( f \)-electrons delocalization [15,22].

In Ref. [24], quantum oscillations were observed in the \( C_T \) mode only, and only between \( B_m \) and \( B^* \). In our experiments, on the contrary, we observed magnetoacoustic quantum oscillations in all modes both in static and pulsed-field measurements. Furthermore, the oscillations are the strongest in the \( C_{33} \) mode.

Figure 9(a) shows the field dependence of the ultrasound-velocity variation in the \( C_{33} \) mode measured in pulsed magnetic fields. Since the anomalies at \( B_m \) and \( B^* \) are not observed in this mode, we also show a similar curve in the \( C_T \) mode as a reference. The oscillations with frequency \( A \) are clearly visible in \( C_{33} \), both below and above \( B^* \), even without subtracting a background. The presence of the frequency \( A \) above \( B^* \) is confirmed by performing the FFT on the background-subtracted data. The corresponding FFT spectra below and above \( B^* \) are shown in Fig. 9(b).

Our static-field data in the \( C_{33} \) mode obtained at lower temperatures, an example of which is shown in the inset of Fig. 10, reveal many more quantum-oscillation frequencies. Some of the frequencies are clearly visible at as low field as 5 T. After subtracting a nonoscillating background from the data shown in the inset of Fig. 10, we performed FFTs over small equidistant \( 1/B \) ranges to keep the same frequency resolution. The resulting field dependence of the FFT spectra is shown in Fig. 10, where the field corresponding to the transition at \( B^* \) is highlighted. It is clear from this FFT analysis that no frequencies emerge or disappear above \( B^* \). This is in
agreement with the results of our recent angle-dependent dHvA study [14]. However, some of the dHvA frequencies observed at high fields in our previous study [14] are not observed in our ultrasound-velocity data.

**IV. CONCLUSIONS**

In summary, we performed high-field ultrasound-velocity measurements on bulk single crystals of CeRhIn₅. For a magnetic field slightly tilted away from the c axis, we observed distinct ultrasound-velocity anomalies at both \( B_m \simeq 20 \) T and \( B^* \simeq 30 \) T at low temperatures in all symmetry-breaking modes, i.e., \( C_{11}, C_{44}, C_{66}, \) and \( C_T \). In all these modes, the anomalies are of similar shape but of opposite sign at \( B_m \) and \( B^* \). Both anomalies are absent in the symmetry preserving \( C_{33} \) mode. Furthermore, our temperature-dependent measurements reveal that both anomalies exist within the AFM state only. Given that the transition at \( B_m \) corresponds to a change of magnetic structure from IC below \( B_m \) to C above \( B_m \), we argue that the transition at \( B^* \) is of the same origin, i.e., from the C phase below \( B^* \) to a new IC phase above it. This makes CeRhIn₅ one of the rare compounds, in which the application of a high magnetic field induces a C to IC transition. High-field neutron diffraction measurements would be of interest to definitely confirm our hypothesis.

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