New High Field State of Flux Line Lattice in Unconventional Superconductor CeCoIn$_5$

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Ultrasound velocity measurements of the unconventional superconductor CeCoIn$_5$ with extremely large Pauli paramagnetic susceptibility reveal an unusual structural transformation of the flux line lattice (FLL) in the vicinity of the upper critical field. The transition field coincides with that at which heat capacity measurements reveal a second order phase transition. The lowering of the sound velocity at the transition is consistent with the collapse of the FLL tilt modulus and a crossover to quasi two-dimensional FLL pinning. These results provide a strong evidence that the high field state is the Fulde-Ferrell-Larkin-Ovchinikov phase, in which the order parameter is spatially modulated and has planar nodes aligned perpendicular to the vortices.

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In almost all superconductors, once the energy gap in the electronic spectrum opens at the critical temperature $T_c$, only the gap amplitude, but not the shape and symmetry around the Fermi surface, changes within the SC state. The only exceptions to this well-known robustness of the SC gap symmetry have, until now, been reported for UPt$_3$[1], Sr$_2$RuO$_4$[2], PrOs$_4$Sb$_{12}$[3], as well as for superfluid $^3$He. In these materials, multiple SC phases with different symmetries manifest themselves below $T_c$. Moreover, in these superconductors the SC order parameter possesses a multiplicity, with a near-degeneracy of order parameters with different symmetries. Tuning the pairing interaction by an external perturbation such as a magnetic field then causes a SC state of given symmetry to undergo a transition to a different SC state.

On the other hand, in the early 1960’s, Fulde and Ferrell and Larkin and Ovchinnikov (FFLO) have developed theories[4], different from the above-mentioned mechanism, for a novel SC phase with a different SC gap function. Generally, in spin singlet superconductors, superconductivity is suppressed by a magnetic field as a consequence of its coupling to the conduction electron spins (Pauli paramagnetism) or to the orbital angular momentum (vortices), both of which break up the Cooper pairs. The FFLO phase appears when Pauli pair-breaking dominates over the orbital effect[5–9]. In the FFLO state, pair-breaking arising from the Pauli effect is reduced by forming a new pairing state $(k^\uparrow, -k + q\downarrow)$ with $|q| \sim 2\mu_B H/\hbar v_F$ ($v_F$ is the Fermi velocity) between exchange-split parts of the Fermi surface, instead of $(k^\uparrow, -k^\downarrow)$-pairing in ordinary superconductors. As a result, a new SC state with different order parameter appears in the vicinity of the upper critical field $H_{c2}$. One of the most fascinating aspects of the FFLO state is that Cooper pairs have a drift velocity in the direction of the applied field, and develop a spatially modulated order parameter and spin polarization with a wave length of the order of $2\pi/|q|$. In spite of enormous efforts to find the FFLO state, it has never been observed in conventional superconductors. The question of observing the FFLO phase in an unconventional superconductor has only been addressed more recently. In the last decade, heavy fermion superconductors CeRu$_2$ and UPd$_2$Al$_3$ have been proposed as candidates for the observation of the FFLO state, but subsequent research has called the interpretation of the data for these materials in terms of an FFLO state into question[10].

Recently it was reported that CeCoIn$_5$ is a new type of heavy fermion superconductor with quasi two dimensional (2D) electronic structure and an effective electron mass $m^* \approx 100m_e$[11]. CeCoIn$_5$ has the highest transition temperature ($T_c$=2.3 K) of all heavy fermion superconductors discovered until now. Subsequent measurements have identified CeCoIn$_5$ as an unconventional superconductor with, most likely, d-wave gap symmetry[12, 13]. Very recent reports of heat capacity measurements in field parallel to the ab-plane of CeCoIn$_5$ have raised great interest, because they possibly point to the occurrence of a FFLO phase[14, 15]. The phase transition from SC to normal metal at the upper critical field parallel to ab-plane, $H^\parallel_{c2}$, is of first order below approximately 1.3 K, in contrast to the second order transition in other superconductors[13–16]. A second order phase transition line, which branches from the first order $H^\parallel_{c2}$-line and decreases with decreasing $T$, was observed within the SC state below 0.35 K[14]. CeCoIn$_5$ satisfies the requirements for the formation of FFLO state. First, it is in the very clean regime, i.e. the quasiparticle mean free path is much larger than the coherence length $\xi$. Second, the Ginzburg-Landau parameter $\kappa = \lambda/\xi \gg 10$, with $\lambda$ the penetration length, is very large. Thirdly, $H^\parallel_{c2}(T=0)$ has an extremely high value of 11.9 T, obeying to a very large conduction electron mass and to the two dimensionality. This situation is favorable for the occurrence of the FFLO state because
the Pauli effect may overcome the orbital effect. Fourth, the pairing symmetry is most likely to be $d$-wave, which greatly extends the stability of the FFLO state with respect to a conventional superconductor \cite{8}. On the basis of some of these arguments, the possibility of a high-field FFLO state has been evoked \cite{14, 15}. However, there still is no corroborated evidence for the FFLO state, because of other possible origins for a second order transition, such as some sort of magnetic transition in the heavy fermion system. Therefore, a more detailed experimental investigation of the vortex state is required in order to shed light on this subject.

The most salient feature of the FFLO state predicted by a number of theories is that the SC order parameter has planar nodes aligned perpendicularly to the applied field. In particular, as pointed out in Ref. 6, the flux lines are divided into segments with distance $\sim 2\pi/|\mathbf{q}|$. Therefore, in order to establish the existence of a possible FFLO state, it is particularly important to elucidate the structure of the flux line lattice (FLL), which in turn is intimately related to the electronic structure. Here, we present ultrasound velocity measurements which provide direct information on the FLL structure in CeCoIn$_5$. In ultrasound experiments, sound waves are coupled to the FLL when the latter is pinned by crystal lattice defects. The ensuing modified sound dispersion allows one to extract detailed information about the FLL-crystal coupling. We find a distinct anomaly of the sound velocity, the nature and magnitude of which provides strong evidence for a vortex state with modulated order parameter as predicted for the FFLO phase.

The ultrasound velocity measurements were performed on high quality single crystals of CeCoIn$_5$ with tetragonal symmetry, grown by the self-flux method. Ultrasonic waves, with a frequency of 90 MHz, were generated by a LiNbO$_3$ transducer glued on the surface of the crystal. A pulse echo method with a phase comparison technique was used for the sound velocity measurements. The relative change of the sound velocity was measured by the phase change of the detected signal. The resolution of the relative velocity measurements was about 1 part in $10^6$. In all measurements, the magnetic field $\mathbf{H}$ was applied parallel to the 2D planes ($\mathbf{H} || [100]$). The inset to Fig. 1(b) shows $v_0^0$ in zero field, and in the normal state above $H_{c2}^\parallel$. In zero field, $v_0^0$ shows a steep increase below $T_c$, indicating a stiffening of the crystal lattice at the superconducting transition. In all experiments presented here, the crystal lattice response is limited by its shear modulus, $C_{66}$.

Experiments on the FLL were always carried out in the field-cooled condition. Figures 1 (a) and 1(b) display the transverse sound velocity $v_0^t$ as a function of temperature in two different configurations with respect to the polarization vector $\mathbf{u}$, the sound propagation vector $\mathbf{k}$, and $\mathbf{H}$. We distinguish between the Lorentz force mode (Fig. 1 (a), $\mathbf{u} \perp \mathbf{H}$) where the sound wave couples to the vortices through the induced Lorentz force $\mathbf{F}_L \approx \lambda^{-2}(\mathbf{u} \times \mathbf{H}) \times \mathbf{B}$, and the non-Lorentz mode (Fig. 1 (b), $\mathbf{u} || \mathbf{H}$) where the sound wave couples only to the crystal lattice. As shown

![Fig. 1: The transverse sound velocity $v_0^t$ as a function of temperature in two different configurations: (a) the Lorentz mode $\mathbf{H} || \mathbf{k} || [100]$, $\mathbf{u} || [010]$ and (b) the non-Lorentz-mode, $\mathbf{u} || \mathbf{H} || [100]$, $\mathbf{k} || [010]$. The configuration (a) corresponds to a flux line bending mode. The inset to Fig. 1(b) shows $v_0^0$ in zero field and in the normal state above $H_{c2}^\parallel$.](image1)

![Fig. 2: The relative shift of the transverse sound velocity $\Delta v_0^t/v_0^t$ as function of $H$ at 100 mK for the Lorentz ($\mathbf{H} \perp \mathbf{u}$) and the non-Lorentz modes ($\mathbf{H} \parallel \mathbf{u}$). The inset depicts the difference between Lorentz and non-Lorentz modes, $\Delta v_0^t = v_0^0(\mathbf{u} \perp \mathbf{H}) - v_0^0(\mathbf{u} \parallel \mathbf{H})$, which can be attributed to the contribution of the FLL. The dashed line shows a fit to Eq. (1).](image2)
in Figs. 1 (a) and (b) and Fig. 2, the transverse sound velocity in the Lorentz mode, in which flux motion is parallel to the ab-plane, is strongly enhanced compared to that in the non-Lorentz mode. This indicates that the transverse ultrasound strongly couples to the FLL in the Lorentz mode. The difference between the sound velocity in the Lorentz mode and in the non-Lorentz mode, \( \Delta v_t = v_{\parallel}\!(t \perp H) - v_{\parallel}\!(t \parallel H) \), can be regarded as being the contribution of the FLL to the sound velocity. In the Lorentz mode, Fig. 1(a), the transverse polarization of the sound has the FLL undergo a long-wavelength (|\( \mathbf{k} \)| \( \lambda \ll 1 \) ) bending mode, limited by the FLL tilt modulus in the limit |\( \mathbf{k} \)| \( \rightarrow 0 \). This particular origin of the transition. This can be seen by looking at \( \Delta v_t/v_t \) and \( \Delta v_{\parallel}/v_{\parallel} \) respectively for the Lorentz mode sound velocities. For clarity, the zero levels of the different curves are vertically shifted. At \( T^* \) marked by arrows, \( \Delta v_t/v_t \) exhibits a distinct cusp. The dashed lines are \( \Delta v_t/v_t \) extrapolated from above \( T^* \). Dotted arrows indicate \( T_c(H) \).

![FIG. 3: The relative shift of the transverse sound velocity velocity arising from the contribution of the FLL, \( \Delta v_t/v_t \), as a function of temperature, below the upper critical field \( H_{c2} \). \( \Delta v_t/v_t \) is obtained as the difference between the non-Lorentz mode and the Lorentz mode sound velocities. For clarity, the zero levels of the different curves are vertically shifted. At \( T^* \) marked by arrows, \( \Delta v_t/v_t \) exhibits a distinct cusp. The dashed lines are \( \Delta v_t/v_t \) extrapolated from above \( T^* \). Dotted arrows indicate \( T_c(H) \).](image)

with \( \rho \) the mass density of the superconducting material, \( j_c \) its critical current density, and \( \Phi_0 = h/2e \).

Figure 2 shows the relative shift of the transverse sound velocity \( \Delta v_t/v_t \) as a function of field at very low temperature. The hysteresis arising from the trapped field was very small. Below approximately 9 T, \( \Delta v_t/v_t \) for the non-Lorentz mode is nearly \( H \)-independent while \( \Delta v_t/v_t \) increases steeply with \( H \) for the Lorentz mode. By fitting the increase to Eq. (1), we obtain a (field–dependent) critical current density, \( j_c = 7.0 \times 10^6 \text{Am}^{-2} \). In both modes, \( \Delta v_t/v_t \) decreases gradually at higher field with \( H \), followed by a jumplike decrease at \( H_{c2} \). This sharp drop, which occurs in a very narrow \( H \)-range \( \Delta H/H_{c2} < 1\% \), is an indication of the first order transition, and disappears at high temperature where the transition becomes second order.

In Figure 3, several curves of the relative shift of the FLL contribution \( \Delta v_t/v_t \) below \( H_{c2} \) are shown as a function of temperature. As the temperature is lowered below the transition temperature in magnetic field \( T_c(H) \), \( \Delta v_t/v_t \) starts to increase. At fields of 9.5 T and higher, \( \Delta v_t/v_t \) changes its slope with a distinct cusp at \( T^* \), as marked by the arrows, while at \( H = 8.5 \text{T} \), no anomaly is observed. Obviously, \( T^* \) separates two different vortex states with different transverse sound velocities: \( \Delta v_t/v_t \) below \( T^* \) is smaller than the \( \Delta v_t/v_t \) extrapolated from above \( T^* \). This indicates a sudden decrease of FLL coupling to the crystal lattice below \( T^* \). Such an anomaly in the ultrasound velocity was not observed when the magnetic field was swept, probably because it occurs near the maximum of \( \Delta v_t(H) \).

Figure 4 displays the \( H-T \) phase diagram below 0.7 K. In this temperature range, the transition at \( H_{c2} \) is of first order as indicated by the sharp drop of the ultrasound velocity (see Fig. 2). In the same figure, we plot the \( H_{c2} \)-line reported in Ref. 14. Although \( H_{c2} \) in our crystal is slightly higher than \( H_{c2} \) reported in Ref. 14, \( T^* \) seems to coincide well with the reported second order transition line[14, 15]. On the basis of our results, we conclude that the second order phase transition is characterized by a sudden change of flux line pinning, due to a structural transition of the FLL. The present results definitely rule out the possibility that some kind of magnetic transition, or a structural phase transition of the crystal lattice, is at the origin of the transition. This can be seen by looking at \( \Delta v_t/v_t \) in the non-Lorentz modes (Fig. 1 (b)), which show no anomaly at \( T^* \).

We here discuss several possible origins for a structural change of the FLL. A reduction of the ultrasound velocity with \( H \) at the transition would occur when the vortex lattice melts into a liquid, as observed in layered high-\( T_c \) cuprates and amorphous thin films[18]. However we can completely rule out this scenario, because the entropy jump at \( T^* \) reported in Ref. 14, 15 is significantly larger than that expected for vortex lattice melting. In addition, flux pinning vanishes at a FLL melting transition, causing \( \Delta v_t/v_t \) to abruptly drop to zero, at odds with the experimental observation. A second possibility is a FLL order-disorder transition in the presence of pinning. However, this should be accompanied by an abrupt increase of the measured critical current and of the ultrasound velocity, whereas the present data show a decrease. Such an effect has been discussed in the light of a possible FFLO state in CeRu2 and UPd2Al3[19]. Thirdly, there might be a possibility that the nature of FLL pinning changes above the transition. For instance, new pinning
anomalies in the non-Lorentz mode, as discussed before. This effect may cause the change of the sound velocities, such as localized spins, could be induced by the transition, and solid line depict the upper critical field $H_{c2}$ determined by the ultrasound experiments. In this temperature regime, the transition at $H_{c2}^\parallel$ is of first order. The dash-dotted line is the second order transition line determined by the heat capacity reported in Ref. [14]. The region shown by green is in the FFLO state. The schematic figures are sketches of a flux line above and below the transition.

The failure of all the above scenarios leads us to consider the possibility that the anomaly in the FLL pinning force and the sound velocity is intimately related to a change of the quasiparticle structure in the high field SC phase, with a reduced electron correlation along the direction of the FLL in the high field phase. Starting from the hypothesis of a transition from a "continuous" superconductor to one with an order parameter structure, modulated in the field direction, with no electronic correlations between layers, we can understand the modest decrease of $\Delta v_t/v_t$. From the critical current density above $T^*$, we deduce the pinning strength $W$ of crystalline defects such as small dislocation loops or stacking faults. For a three-dimensional superconductor, this should be evaluated in the single vortex limit[21]:

$$W = (\Phi_0^{7/2} B^{3/2} j_c^3/4\pi^2 \mu_0 \lambda_c^2)^{1/2} = 5 \times 10^{-4} \text{N}^2 \text{m}^3.$$  

At the transition to the phase with modulated order parameter, the vortex tilt stiffness in the limit of the small scale distortions caused by pinning collapses. Then, the critical current is given by the expression for a 2D layer of thickness $d$, $j_{c2}^{2D} = W/\Phi_0^{7/2} B L_G d$. At the transition, $j_{c2}^{2D}$ must equal to the experimental value $7 \times 10^5 \text{Am}^{-2}$, an equality, that, given the experimental value of $W$, is satisfied for a layer thickness of $4 \times 10^{-8} \text{m}$. This is remarkably close to the modulation $2\pi/|q| = (m_u/m^*) (\hbar k_F/eB) = 3.5 \times 10^{-8} \text{m}$ expected for the order parameter structure associated with the formation of the FFLO state proposed by Ref. 5–9 (the Fermi wavevector $k_F \approx 8 \times 10^4 \text{m}^{-1}$). In the FFLO state, the order parameter performs one-dimensional spatial modulations with a wavelength of the order of $2\pi/|q|$, which is comparable to $\lambda$, along the magnetic field, forming planar nodes that are periodically aligned perpendicularly to the flux lines. The occurrence of planar nodes leads to a segmentation of the flux lines. A schematic figure of this state is shown in Fig. 4. This field-induced layered structure resembles the vortex state of layered cuprates and organic superconductors in magnetic field perpendicular to the conducting planes. The segmentation of vortex lines to a length $2\pi/|q|$ much smaller than the characteristic “Larkin length” $L_c = (c_{el} B/\Phi_0 W^{1/2})^{2/3}$ on which they are pinned, means that the vortex part of the ultrasound velocity is now governed by the pinning of the quasi 2D vortex layers of thickness $2\pi/|q|$. Note that a decrease of $\Delta v_t/v_t$ at the transition can only be obtained by a true dimensionality change of the FLL at the transition, and not by a mere softening of the FLL elastic constants.

In summary, ultrasound velocity measurements of CeCoIn$_5$ in the vicinity of the upper critical field $H_{c2}$ reveal an unusual structural transformation of the FLL at the second order phase transition within the SC state. This transformation is most likely characterized by the collapse of the FLL tilt modulus and a transition to a quasi-2D state. These results are a strong indication that the high field superconducting state is the FFLO phase, in which the order parameter is spatially modulated and has planar nodes aligned perpendicularly to the FLL.

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