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High-field state of the flux-line lattice in the 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 vortex segmentation and a crossover to quasi-two-dimensional FLL pinning. These results provide strong evidence that the high-field state is the Fulde-Ferrel-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_2RuO_4,[2] $PrOs_4Sb_{12}$,[3] as well as for superfluid ^3He. 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 1960s, Fulde and Ferrel and Larkin and Ovchinikov (FFLO) developed theories,[4] different from the above-mentioned mechanism, for an 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 $(\boldsymbol{k}\uparrow, -\boldsymbol{k}+\boldsymbol{q}\downarrow)$ with $|\boldsymbol{q}| \sim 2\mu_B H/\hbar v_F$ (v_F is the Fermi velocity) between exchange-split parts of the Fermi surface, instead of $(\boldsymbol{k}\uparrow, -\boldsymbol{k}\downarrow)$-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 wavelength of the order of $2\pi/|\mathbf{q}|$. 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_2Al_3 have been proposed as candidates for the observation of the FFLO state, but subsequent research has called into question the interpretation of the data for these materials in terms of a FFLO state.[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 $m^* \approx 100 m_e$.[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 heat capacity measurements in field parallel to the ab plane of $CeCoIn_5$ have raised great interest, because they may 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, $H_{c2}^{\parallel}$, is of first order below approximately 1.3 K, in contrast to the second-order transition in other superconductors.[13–16] Within the SC state, a second-order phase transition line branches from the first-order $H_{c2}^{\parallel}$ line below 0.35 K; the transition field decreases with decreasing T.[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 ξ. Second, the Ginzburg-Landau parameter $\kappa = \lambda/\xi \gg 10$ is very large (λ is the penetration depth). Third, $H_{c2}^{\parallel}(T=0)$ has an extremely high value of 11.9 T, owing to a

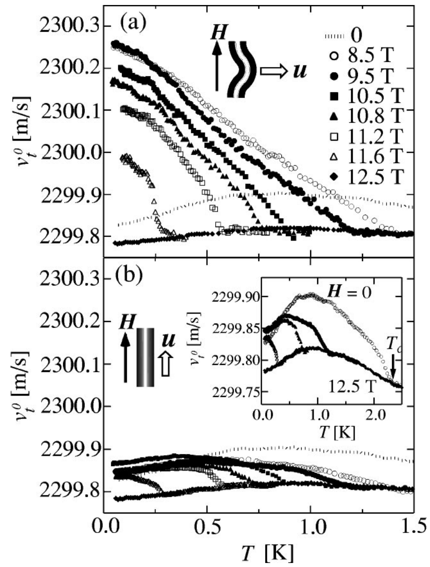

FIG. 1. The transverse sound velocity v_t^0 as a function of temperature in two different configurations. (a) The Lorentz mode $\mathbf{H}\|\mathbf{k}\|[100]$, $\mathbf{u}\|[010]$ and (b) the non-Lorentz mode, $\mathbf{u}\|\mathbf{H}\|[100]$, $\mathbf{k}\|[010]$. The configuration (a) corresponds to a flux line bending mode. The inset to (b) shows v_t^0 in zero field and in the normal state above $H_{c2}^{\|}$, including the high-field data in the non-Lorentz mode with $H<H_{c2}^{\|}$ (9.5 T, 10.8 T, and 11.6 T from right to left).

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.[8] On the basis of some of these arguments, the occurrence of a high-field FFLO state has been evoked.[14,15] However, the transition[14,15] may well be of completely different nature, such as, e.g., a magnetic transition in the heavy fermion system. Therefore, an experimental probe sensitive to the nature of the high field SC state is required in order to corroborate its interpretation in terms of the FFLO phase.

The most salient feature of the FFLO state is that the SC order parameter should have planar nodes aligned perpendicular to the applied field. In particular, magnetic flux lines are expected to be divided into segments of length $\sim 2\pi/|\mathbf{q}|$.[6] Therefore, in order to establish the existence of a possible FFLO state, it is 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 $CeCoIn_5$. 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 segmentation of vortices arising from the modulated order parameter predicted for the FFLO phase.

The measurements were performed on high quality single crystals of $CeCoIn_5$ with tetragonal symmetry, grown by

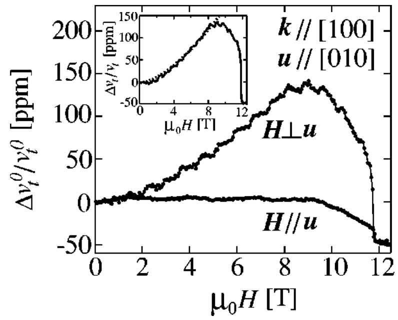

FIG. 2. The relative shift of the transverse sound velocity $\Delta v_t^0/v_t^0$ as function of H at 100 mK for the Lorentz ($\mathbf{H}\perp\mathbf{u}$) and the non-Lorentz modes ($\mathbf{H}\|\mathbf{u}$). The crystal stiffening at T_c results in a negative apparent $\Delta v_t^0/v_t^0$ for $H>H_{c2}$. The inset depicts the difference between Lorentz and non-Lorentz modes, $\Delta v_t = v_t^0(\mathbf{u}\perp\mathbf{H}) - v_t^0(\mathbf{u}\|\mathbf{H})$, which can be attributed to the FLL. The dashed line is a fit to Eq. (1).

the self-flux method. The crystal has rectangular shape with the size of $1.8\times0.7\times0.5$ mm^3. Transverse 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 to measure the relative change of the sound velocity through 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 two-dimensional planes ($\mathbf{H}\|[100]$). The inset to Fig. 1(b) shows v_t^0 in zero field, and in the normal state above $H_{c2}^{\|}$. In zero field, v_t^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 transverse polarization of the sound means that the crystal lattice response is limited by its shear modulus, C_{66}^c.

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_t^0 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 in Figs. 1 and 2, the transverse sound velocity in the Lorentz mode, with flux motion 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, $\Delta v_t = v_t^0(\mathbf{u}\perp\mathbf{H}) - v_t^0(\mathbf{u}\|\mathbf{H})$, 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 $|\mathbf{k}|\to 0$, $c_{44}^f(|\mathbf{k}|\to 0) = B^2/\mu_0$. In the present case of sufficiently strong FLL pinning, the contribution of viscous drag and

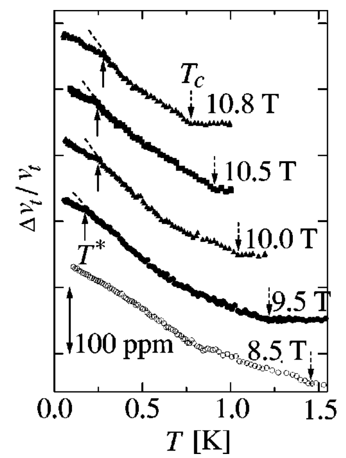

FIG. 3. The relative shift of the transverse sound velocity arising from the contribution of the FLL, $\Delta v_t/v_t$, as a function of temperature, below the upper critical field $H_{c2}^{\parallel}$. $\Delta v_t/v_t$ is obtained as the difference between the non-Lorentz mode and the Lorentz mode sound 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 $T_c(H)$.

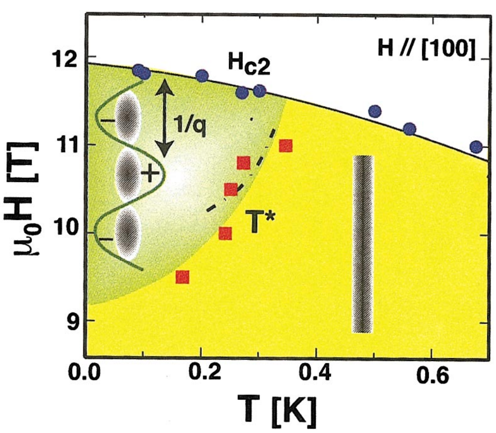

FIG. 4. (Color) Experimental $H-T$ phase diagram for $CeCoIn_5$ below 0.7 K for ($\mathbf{H}\|[100]$). The transition temperature T^*, indicated by red squares, was obtained from ultrasonic measurements as indicated by the arrows in Fig. 3. Blue circles and solid line depict the upper critical field H_{c2} determined by the ultrasonic 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.

normal excitations can be neglected. The FLL contribution then writes[17]

$$\Delta v_t = \frac{B^2}{\mu_0 \rho v_t} \frac{1}{1 + (\Phi_0 B)^{1/2}|\mathbf{k}|^2/\mu_0 j_c}, \tag{1}$$

with ρ 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 $\Delta v_t^0/v_t^0$ as a function of field at very low temperature. The hysteresis arising from the trapped field was very small. Below approximately 9 T, $\Delta v_t^0/v_t^0$ for the non-Lorentz mode is nearly H-independent while $\Delta v_t^0/v_t^0$ increases steeply with H for the Lorentz mode. By fitting the increase to Eq. (1), we obtain a (field-independent) critical current density, $j_c = 7.0 \times 10^8$ A m^{-2}. In both modes, $\Delta v_t^0/v_t^0$ decreases gradually at higher field with H, followed by a jumplike decrease at $H_{c2}^{\parallel}$. This sharp drop, which occurs in a very narrow H-range $\Delta H/H_{c2}^{\parallel} < 1\%$, is an indication of the first-order transition, and disappears at high temperature where the transition becomes second order.

In Fig. 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 $H = 8.5$ 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 marked decrease of FLL coupling to the crystal lattice below T^*. The anomaly in the ultrasound velocity was not easily resolved when the magnetic field was swept, because it occurs in the field regime near H_{c2} in which the overall $\Delta v_t(H)$ decreases as a result of vortex core overlap.

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 $H_{c2}^{\parallel}$ in our crystal is slightly higher than $H_{c2}^{\parallel}$ 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 $\Delta v_t^0/v_t^0$ in the non-Lorentz modes [Fig. 1(b)], which show no anomaly at T^*.[18]

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.[19] However we can completely rule out this scenario, because the entropy associated with the transition reported in Refs. 14 and 15, $20k_B$ per vortex line segment of length ξ, 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 $CeRu_2$ and UPd_2Al_3.[20]

Third, one can hypothesize that the nature of FLL pinning changes above the transition. For instance, new pinning centers, such as localized spins, could be induced by the transition, or the vortex core structure may change. However, this would also lead to an increase in $\Delta v_t/v_t$ non-Lorentz mode. Finally, it may be that the FLL undergoes a symmetry change, e.g., from hexagonal to square, with a concomitant lowering of the vortex lattice shear modulus c_{66}^f and increase of the pinning force. Such a transition has indeed been observed in $CeCoIn_5$,[21] but at $\mu_0 H = 0.6$ T, much lower than the fields involved here.

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, 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:[22] $W = (\Phi_0^{7/2} B^{3/2} j_c^3 / 4\pi^2 \mu_0 \lambda^2)^{1/2} = 5 \times 10^{-4}$ $N^2 m^3$. This value implies that vortex excursions along the field direction are correlated on the Larkin length, $L_c = (c_{44}^f \Phi_0 / B W^{1/2})^{2/3} \approx 1$ μm. At the transition to the phase with modulated order parameter, the modulation length $2\pi/|\mathbf{q}|$ starts to decrease. This decrease does not affect the FLL structure and the j_c until $2\pi/|\mathbf{q}| \approx L_c/2$.[22] At this point, the vortex tilt modulus becomes irrelevant and the critical current is given by the expression for a 2D layer of thickness d, $j_c^{2D} = W/\Phi_0^{1/2} B^{1/2} c_{66}^f d$. Given the value of W, j_c^{2D} turns out to equal the experimental value 7×10^8 A m^{-2} for a layer thickness of 4×10^{-8} m. This is remarkably close to the modulation $2\pi/|\mathbf{q}| = (m_e/m^*)(hk_F/eB) = 3.5 \times 10^{-8}$ m expected for the order parameter structure associated with the formation of the FFLO state proposed by Refs. 5–9 (the Fermi wave vector $k_F \approx 8 \times 10^9$ m^{-1}). In the FFLO state, the order parameter performs one-dimensional spatial modulations with a wavelength of the order of $2\pi/|\mathbf{q}|$, which is comparable to ξ, along the magnetic field, forming planar nodes that are periodically aligned perpendicular 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 smaller than the Larkin length means that the vortex part of the ultrasound velocity is now governed by the pinning of quasi-2D vortex layers with $d = 2\pi/|\mathbf{q}|$. Note that the *decrease* of $\Delta v_t/v_t$ at the transition can only be obtained by the dimensionality change descibed above, and can only happen following a *continuous* increase of the $|\mathbf{q}|$. A discontinuous increase, or a gradual increase of the modulation *amplitude* would have resulted in a jump of $\Delta v_t/v_t$.

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 vortex segmentation, i.e., as 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 perpendicular to the FLL.

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