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Critical Field and Shubnikov-de Haas Oscillations of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ under Pressure

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We have used a novel experimental method to study the crossover of an anisotropic superconductor from a possible Pauli limited superconducting state to a vortex limited superconducting state by applying pressure. The new apparatus combined a tuned tank circuit with a nonmetallic diamond anvil cell to measure the change in critical field with angle in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ at pressures up to 1.75 kbar and at temperatures down to 70 mK. The critical fields (in the perpendicular or parallel orientation to the conducting planes) have been found to decrease by more than 90% within less than 2 kbar of pressure. In the parallel orientation, at 1.75 kbar, we have seen a clear change from the ambient pressure behavior of the critical field with temperature at low temperatures. Up to $P = 1.75$ kbar, the $H_{c2}(\theta)$ phase diagram is in good agreement with the theoretical prediction for weakly coupled layered superconductors. We have also succeeded in measuring oscillations in the resistivity of the normal state at higher magnetic field which could be used to find the effective quasi-particle mass. The α-orbit Shubnikov-de Haas frequency was found to increase at a rate of 44 T/kbar. Our experiment opens the possibility for further investigations of the effective mass with pressure, especially because the setup is suitable for pulsed fields as well.

***KEY WORDS*:** Shubnikov-de Haas Oscillations; FFLO state; Pauli limited.

1. INTRODUCTION

A number of anisotropic and heavy fermion superconductors have recently been found to exhibit properties associated with an inhomogeneous superconducting order parameter.[1–4] Similar novel superconducting states were first predicted by Fulde and Ferrell and Larkin and Ovchinnikov[5,6] and are often called FFLO states. The FFLO state is expected in clean superconductors that are Pauli limited. We call a superconductor Pauli limited when the interaction of the applied magnetic field with the electron spin limits the superconducting state, in contrast to orbital limiting, the traditional effect where vortices eventually destroy the superconducting state. One way to favor Pauli limiting over orbital limiting is to create a layered material and orient the applied magnetic field along the layers. In this orientation the electrons have to tunnel across the insulating layers to form vortices, and it becomes energetically favorable for the applied magnetic field lines to pass between the conducting layers and form weak Josephson vortices. Our motivation for this experiment was to increase the pressure on a highly anisotropic, quasi 2D superconductor that shows evidence of Pauli limiting, and by applying this pressure, increase the interlayer conduction and change the dimensionality of the superconductor so that orbital limiting would become dominant.

Due to its strong two-dimensional character, the charge-transfer organic salt κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is a suitable material for studying the various theories put forth for anisotropic superconductivity in magnetic fields. The electronic structure of organic superconductors is very similar to that of the cuprate high T_c superconductors, consisting of stacks of alternating conducting and insulating sheets. In contrast to the cuprates, however, the critical fields of organic superconductors are much lower making them easier to study. Among the organics, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ with a $T_c = 10.4\,\mathrm{K}$, has been shown to be one of the compounds with the highest critical fields. Even with its conducting planes parallel to the applied field, H_{c2} is less than 40 tesla (T).[7,8] Furthermore, high-purity single crystals of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ are available which make for reliable studies of the Fermi surface. For example, the samples used in this study have mean free paths from 600 to 900 Å, and a superconducting coherence length in the layers of ~ 100 Å. These parameters put κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ clearly in the clean limit. YBCO in comparison has a mean free path less than 100 Å, and a superconducting coherence length in the layers of ~ 50 Å.[9]

The strong effect of the pressure on the band structure via modification of the carrier effective mass and Fermi surface (and hence on the superconducting properties) already reported in the literature[10] has motivated our work. In the present paper, we focus on the change in critical field in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ under pressure for different orientations of the applied dc magnetic field with respect to the conducting planes (we will refer to θ as the angle between the magnetic field and the normal to the conducting planes). The study of reduced dimensional systems is important, because of the different mechanisms which destroy the superconductivity when the magnetic field is applied perpendicular or parallel to the conducting layers[11] as described above. In the absence of any other mechanisms (spin-orbit scattering, many body effects), the maximum critical field in the parallel orientation, called the Pauli paramagnetic limit, H_P, is driven by the spin polarization effect, where the condensation energy is overcome by the Zeeman splitting energy.[12,13] In this limit, H_{c2} may change from a second-order to a first-order transition,[14] or an inhomogenious superconducting state may be stabilized.[3,5,6] For this paper we will use the BCS approximation of the Pauli limit, $\mu_0 H_P^{BCS} = \sqrt{2}\Delta_0/g\mu_B$, where the energy gap, $2\Delta_0 = 3.5 k_B T_c$, k_B is Boltzmann's constant, g is the gyromagnetic ratio, T_c is in kelvin, and $\mu_0 H_P^{BCS}$ is in tesla. There may be better ways to calculate the the Pauli limit,[15] but the BCS method will suffice to compare the same material at different pressures. For a comprehensive summary of organic superconductors in the Pauli paramagnetic limit region we refer the reader to Ref. 8.

κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ has been found to have an unusual evolution of the parallel critical field with temperature. In spite of their differences in the measured values of the critical field and in the curvature of the phase diagram at higher temperatures, all the experimental results at ambient pressure agree that the absolute value of $H_{c2}^{||}$ not only exceeds H_P^{BCS}, but it also shows no tendency of saturation at low temperatures[3,16,17] and displays positive curvature toward 0 K, indicating that Pauli effects are present. However, the reason for this behavior is not well understood. We cite two recent hypotheses, one that explains the lack of saturation as a first order phase transition into the FFLO state[3] and another one that claims the high critical fields are indeed beyond the BCS Pauli paramagnetic limit, but comparable to the paramagnetic limit calculated from thermodynamic quantities.[16] Both references seem to ignore the spin-orbit scattering effect, which also can be responsible for the enhancement of the upper critical field. We, however, suggest that spin-orbit scattering cannot be responsible for the total enhancement of the critical field. If this were the case, following Ref. 18, we calculated that the spin-orbit scattering time would be between 0.46 and 0.62 ps, which is much less than the total measured scattering time of about 3 ps determined from magnetoresistance oscillations.[19] Therefore, we can rule out a large role for spin-orbit scattering. It has been predicted that in the absence of spin-orbital scattering, the transition from the normal to the superconducting state at low temperatures should turn into a first-order phase transition.[14] We have found evidence that there is a first order transition below 4 K in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ further indicating that Pauli effects are present.[20] Thus it is valuable to study the effect of the pressure on the $H^{\parallel}_{c2}(T)$ phase diagram and tune the anisotropy, especially because existing data suggests a tendency of saturation of $H^{\parallel}_{c2}$ in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ at 1.5 kbar[21] indicating that the Pauli effects are absent.

At ambient pressure, it is often claimed that κ-(BEDT-TTF)$_2$Cu (NCS)$_2$ is very anisotropic. Although the anisotropy ratio $\gamma = H^{\parallel}_{c2}/H^{\perp}_{c2}$ is only about 6,[16] the anisotropy of the London penetration depths is $\simeq$160–330.[22] We believe the reason why the anisotropy determined by the critical fields is misleading is that the mechanism that limits the superconductivity when the applied field is parallel to the layers is not related to the coherence length, because this critical field is Pauli limited. Hence, the parallel and perpendicular critical fields cannot be used to find the ratio of the parallel and perpendicular coherence lengths, as is common with less anisotropic superconductors. Nevertheless, the $H_{c2}(\theta)$ diagram for κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ fits the Lawrence–Doniach 2D model of weakly coupled layered superconductors despite the Pauli limiting.[16,23] And, as we will show, even under moderate pressure we never see results consistent with anisotropic 3D Ginzburg–Landau theory.[24]

2. EXPERIMENTAL

Our innovation was to combine a nonmetallic diamond anvil cell (DAC)[25] with an rf penetration depth technique.[26] The plastic pressure cell design overcomes the difficulties of using metals in magnetic field, can be made of a relatively small size to fit on a rotating platform and, by placing a ruby chip inside the cell, the pressure can be measured in-situ. The penetration depth was measured using the tunnel diode oscillator (TDO) technique, which offers the advantage of not requiring contacts on the sample, and therefore, eliminates problems like contact resistance and additional stress on the sample. It is particularly well suited for use in the diamond anvil cell because the coil and the sample can be of arbitrarily small size. In a recent advance, we have succeeded in using this combination of techniques in the pulsed field environment to 50 T at He-3 temperatures. The samples were single crystals of κ- (BEDT-TTF)$_2$Cu(NCS)$_2$ approximately 210μm $\times$ 175 μm $\times$ 40 μm. They were placed in a four turn coil (56 AWG wire) with an inner diameter of 300 μm with their conducting planes perpendicular to the axis of the coil. To minimize the background signal, a nonmetallic diamond anvil cell was used with a diamond filled epoxy gasket reinforced by a Zylon overband.[27] The plastic DAC freely rotated in a top-loading dilution refrigerator with an ID of 21.5 mm. The coil rested in the 350 μm diameter hole of the gasket that was filled with the quasi-hydrostatic pressure medium glycerin. Ruby was used to calibrate the pressure at the operating temperature.[28] The TDO setup has been explained in detail elsewhere.[26] The oscillating frequency of the circuit at 70 mK was 290 MHz, and the change in frequency during the sweep of the magnetic field was about 2 MHz, less than 1%. Figure 1 shows typical field dependences of the frequency and amplitude when the field is applied parallel and perpendicular to the conducting layers. The overlap of the inverse amplitude, which is a direct measure of the dissipation in the circuit, and the frequency, attests to the integrity of the

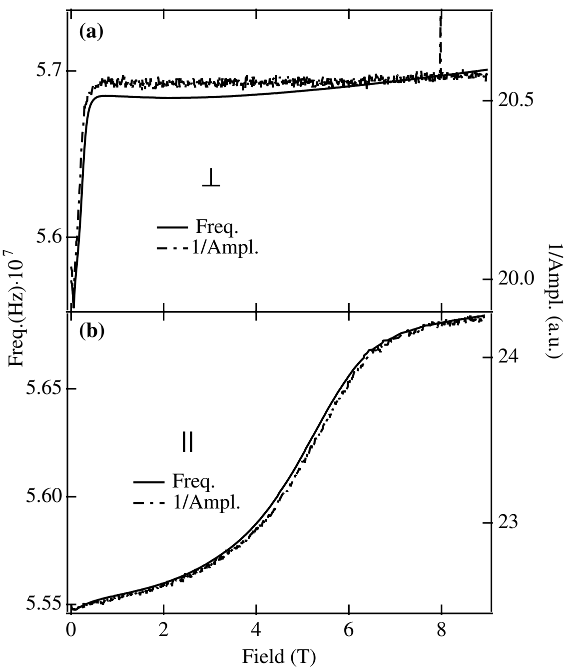

Fig. 1. The rf penetration (proportional to the change in frequency) as a function of magnetic field for the orientation perpendicular to the conducting planes (a) and the parallel orientation (b). H_{c2} is determined by the intersecting point between the linear extrapolation of the normal state and the superconducting transition.

data. We define the critical field as the intersection point between the linear extrapolation of the normal state and the superconducting transition. The data reported in the present work were taken in a top loading dilution refrigerator and an 18 T superconducting magnet system at NHMFL in Tallahassee. The present configuration of the TDO electronics limits the lowest achievable temperature to 70 mK.

3. RESULTS

Figure 2 shows the critical field, both parallel and perpendicular, for ambient pressure and three other values: 1.5, 1.67, and 1.75 kbar at T = 90 mK. The critical fields in the different orientations decrease linearly as the pressure increases. However, the rates of change are different for the two orientations which is consistent with the anisotropy in the critical fields. We found $dH^{\perp}_{c2}/dP \simeq -2.8$ T kbar^{-1} whereas $dH^{\parallel}_{c2}/dP \simeq -14.75$ T kbar^{-1}. Extrapolating the fitting lines, we found a critical pressure P_c of about 1.8 kbar for $H^{\perp}_{c2}$ and 2.1 kbar for $H^{\parallel}_{c2}$. These values are less than half of the value reported in Ref. 10 where $P_c \simeq$ 5 kbar. It is possible that anisotropic stresses of the frozen organic fluid in the pressure cell applied large strains on the sample causing us to underestimate the pressure by using the average value.[29] We present some data to support this idea when we discuss the Fermi surface at the end of Sec. 3 of this paper, although this overestimate cannot account for the full discrepancy of the

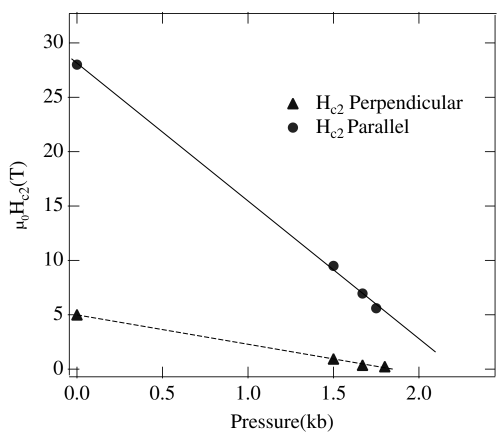

Fig. 2. Pressure dependence of parallel (circles) and perpendicular (triangles) critical field. The error is approximately the size of the symbols. The two ambient pressure points come from Ref. 16.

critical pressures. The suppression of superconductivity by more than 90% within less than 1.5 kbar underlines the importance of a careful study of the effective mass under pressure. However, the very high linear rate of change of the parallel critical field with pressure is also striking, because while the change in the effective mass directly influences orbital effects, we assume that $H_{c2}^{\parallel}$ is not orbitally limited. At this point, we only question the conclusion of Ref.10 that the enhancement of the effective mass is directly associated with superconductivity in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and suggest that other parameters, such as the V_{BCS} interaction term (the electron–phonon coupling matrix element),[30] the density of states and/or the phonon characteristic energy may also be very sensitive to the applied pressure.

We measured the change in critical field with temperature at 1.75 kbar, both in the perpendicular and parallel orientation (Fig. 3). Although both diagrams show a saturation of the critical field at very low temperature,

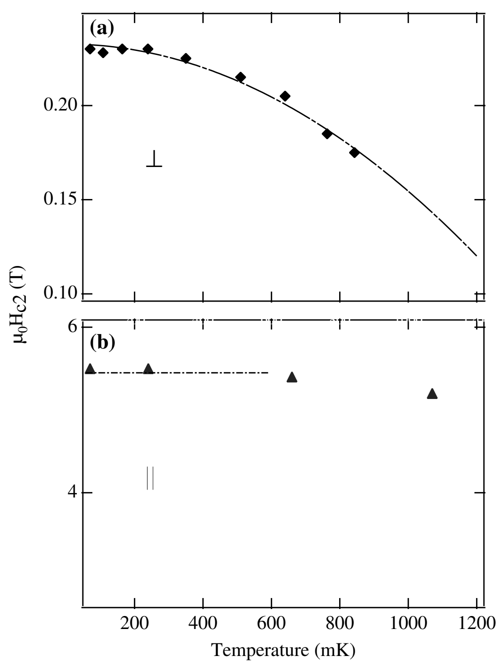

Fig. 3. Critical fields [(a)-$H_{c2}^{\perp}$ and (b)-$H_{c2}^{\parallel}$] as a function of temperature at $P = 1.75$ kbar. The continuous line in (a) is the Ginzburg–Landau equation of the critical field at low temperature: $H_{c2} = \text{Const.} \times (1 - (T/T_c)^2)$. In both graphs, the field axis (y) shows about half of the maximum critical field showing that the parallel field is saturated over much more of the temperature range.

it may not happen for the same reason. In the perpendicular orientation, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is orbitally limited at ambient pressure, the orbital critical field (≈ 5 T) being well below the Pauli limit (≈ 18 T), and we found the same situation at 1.75 kbar. As can be seen in Fig. 3a, our experimental data falls nicely on the theoretical Ginzburg–Landau result, $H_{c2} \approx (1-(T/T_c)^2)$ albeit for only the lower half of the temperature range. A fit and extrapolation yields $T_c \simeq 1.75$ K $\pm$ 0.5 K.

As mentioned in the introduction, ambient pressure studies show that $H^{\|}_{c2}$ exceeds the Pauli paramagnetic limit H_P^{BCS}, and shows no tendency of saturation as $T \rightarrow 0$ K.[11] In contrast, at $P = 1.75$ kbar we found no change in the parallel critical field as the temperature increases from 70 to 240 mK. Above 240 mK it drops with a negative curvature (Fig. 3). Studying how the ratio between the measured $H^{\|}_{c2}$ and the BCS Pauli limit change under pressure, we had to sort out the very different values obtained for the highest $H^{\|}_{c2}$ at ambient pressure. Using the upper critical field of 28 T,[31] the ratio H_{meas}/H_P^{BCS} is 1.55. At 1.75 kbar, if we conservatively estimate T_c to be 2.00 K (from the perpendicular diagram), then H_P^{BCS} would be equal to 3.7 T and the ratio H_{meas}/H_P^{BCS} would be about 1.47. These numbers suggest that the pressure does not change the proportion between the critical field and the energy gap (in so far as the T_c determines the energy gap) and suggests Pauli limiting.

Based on H_{c2} studied in previous experiments, one could expect either an increase or decrease in the parallel critical field as κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is subjected to pressure. If the conducting layers are decoupled and the layers are squeezed, the parallel critical field should increase until the it reaches the Pauli limit, as was found in single layers of aluminum.[32] If the insulating layers are squeezed, the parallel critical field should decrease as the the orbital limiting is enhanced due to the increased coupling of the layers, and the increased perpendicular coherence length. The fact that H_{c2} parallel is saturated at 1.75 kbar and the ratio H_{meas}/H_P^{BCS} does not change suggests that κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is always Pauli limited, and that g has not changed as the pressure is increased. This result is in contrast to the difference in the shape of the critical field phase diagram as the pressure is changed from ambient to 1.75 kbar. The difference in the shape of H_{c2} suggests that there is a fundamental change in the cause of H_{c2}. At all the pressures we worked at up to 1.75 kbar and at temperatures down to 70 mK, we have seen no evidence similar to our experiments in $CeCoIn_5$[15] for the FFLO phase, as was claimed to be present at ambient pressure,[3] and the transition is always of second order (Fig. 1b).

Concluding the above discussion, to understand the mechanism of superconductivity in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ it becomes very important to complete the ambient pressure phase diagram below 500 mK, but it is experimentally difficult to obtain magnetic fields higher than 30–35 T and temperatures below 500 mK at the same time. We have shown that very low pressures, probably less than 1 kbar, would make this experimental investigation much more transparent.

To support our analysis above we would like to make sure that, under pressure, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ does not suffer a transition from a layered quasi 2D superconductor toward an anisotropic 3D superconductor. If that were the case, then the orbital effects would no longer be negligible in the parallel orientation, and could even become the dominant factor. We can experimentally verify this change by measuring the change in the critical field with angle. The Ginzburg–Landau theory for an anisotropic 3D superconductor predicts a variation of the critical with angle after the following equation:[24]

$$\left[\frac{\mathrm{H}_{c2}(\theta)\cos(\theta)}{\mathrm{H}_{c2}^{\perp}}\right]^2 + \left[\frac{\mathrm{H}_{c2}(\theta)\sin(\theta)}{\mathrm{H}_{c2}^{\parallel}}\right]^2 = 1, \tag{1}$$

where θ is the angle between the field and the normal to the layers. For weakly coupled layered superconductors, Tinkham[24] and then Schneider and Schmidt[33] found that the angular dependence is given by:

$$\left|\frac{\mathrm{H}_{c2}(\theta)\cos(\theta)}{\mathrm{H}_{c2}^{\perp}}\right| + \left[\frac{\mathrm{H}_{c2}(\theta)\sin(\theta)}{\mathrm{H}_{c2}^{\parallel}}\right]^2 = 1, \tag{2}$$

which leads to a cusp-like behavior. For a truly Pauli limited superconductor even Eq. (2) should not be valid, because it is still based on orbital destruction of superconductivity. However, one can argue phenomenologically that a similar equation should exist with H_P in place of H_{c2} parallel, which is what we have used in this discussion. A theoretical argument for this phenomenological equation was made by Bulaevskii.[34]

We have determined the $\mathrm{H}_{c2}(\theta)$ diagram for $\mathrm{P} = 1.67$ and 1.75 kbar at $\mathrm{T} = 70$ mK. The experimental result along with the fits by Eqs. (1) and (2), are plotted in Fig. 4. The cusp-like feature observed experimentally at $\theta = 90^\circ$ is the indication that Eq. (2) is a better fit up to 1.75 kbar. Therefore, we confirm experimentally that up to $\mathrm{P} = 1.75$ kbar there is no evidence for moving toward a more 3D (or less 2D) superconductor. We also found an enhancement of the anisotropy in critical field $\gamma = \mathrm{H}_{c2}^{\parallel}/\mathrm{H}_{c2}^{\perp}$ from $\simeq$ 6 at ambient pressure to $\gamma \simeq 23$ at $\mathrm{P} = 1.75$ kbar, but we attribute this increase in the apparent anisotropy to the different mechanisms that affect

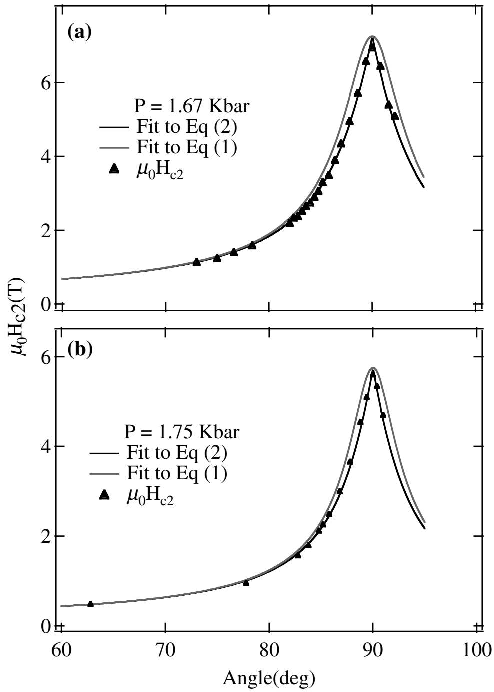

Fig. 4. Angular dependence of H_{c2} at 1.67 kbar (a), and 1.75 kbar (b). For both graphs, the continuous black line represents a fit with Lawrence–Doniach equation and the gray curve is a fit to the anisotropic 3D Ginzburg–Landau equation.

the critical fields in each orientation rather than to an enhancement of the 2D character of the κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, as discussed in the introduction. In reference to our comment earlier that $H_{c2}^{\|}$ at 1.75 kbar is Pauli limited, the cusp like character of the angular dependence is further evidence that the orbital effects are suppressed when the sample is at the parallel orientation. If there was significant transport through the layers, the angular dependence would have a rounded top near 90°.

Beyond the superconducting transition, the change in frequency (and amplitude) of the TDO is due to the resistivity of the normal state, and at higher fields we have measured the Shubnikov-de Haas oscillations in magnetoresistance as shown in Fig. 5. Our limit of 18 T did not allow for a careful analysis of the oscillation frequency with pressure and temperature, but we found an increase in the frequency of α-orbit (F_α) from 694.1 T at 1.5 kbar to 703.0 T at 1.67 kbar (T = 90 mK), while the ambient value (F_0) is about 595 T. (At 1.75 kbar an experimental error prevented us from seeing the SdH oscillations.) The ratio F_α/F_0 is therefore, 1.17 at 1.5 kbar

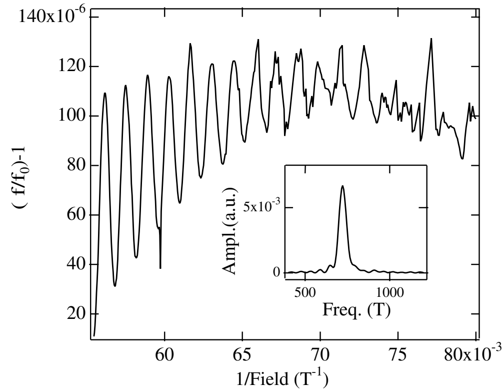

Fig. 5. Magnetoresistance oscillations of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ at P=1.67 kbar and T = 90 mK. The inset shows the FFT of the oscillations.

and 1.18 at 1.67 kbar. The linear increase of the frequency of oscillations with pressure is due to the change in size of the unit cell and the inverse effect on the Brillouin zone.[10,30,35] This frequency corresponds to almost 3 kbar in other experiments using petroleum spirit, and 6 kbar in helium[29] suggesting that non hydrostatic pressure may have caused more stress on the sample than we expected based on our ruby measurements.

4. CONCLUSIONS

In summary, we have proven that the combination of the TDO technique and the nonmetallic pressure cell can provide a very useful tool in the study of superconductivity.

Measurements on κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ revealed that the pressure strongly affects the critical field, by more than 90% within 1.5 kbar, both in the perpendicular and parallel orientations. As far as we have measured, the superconductivity is suppressed linearly as a function of pressure in both orientations.

At 1.75 kbar, we found a clear change in the behavior of the parallel critical field with temperature (non-saturating to saturating), from the ambient pressure phase diagram. Yet, the value of $H^{\parallel}_{c2}$ still exceeds the BCS Pauli limit in the same ratio as at ambient pressure. This conflicting evidence makes it difficult to know if κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is still in the Pauli limit at high pressures, although according to our experimental evidence, up to 1.75 kbar, κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is still well described by the 2D Lawrence–Doniach model for layered superconductors. These points suggest that κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is still Pauli limited even up to 1.75 kbar.

The frequency of the magnetoresistance oscillations increases with pressure at a higher rate than previously reported in literature Ref. 10. In an effort to better understand the role played by different physical quantities (e.g. the effective mass, V_{BCS}, transfer integral, spinorbit scattering rate) we are pursuing larger fields, lower temperatures, and higher pressures. Exploring the very low pressure gap in our data would be very useful as well.

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REFERENCES

1. H. A. Radovan, N. A. Fortune, T. P. Murphy, S. T. Hannahs, E. C. Palm, S. W. Tozer, and D. Hall, *Nature* **425**, 51 (2003).
2. A. Bianchi, R. Movshovich, C. Capan, A. Lacerda, P. G. Pagliuso, and J. L. Sarrao, *Phys. Rev. Lett.* **91**, 187004 (2003).
3. J. Singleton, J. A. Symington, M.-S. Nam, A. Ardavan, and M. Kurmoo, P. Day, *J. Phys.: Condens. Matter* **6**, L641 (2000).
4. M. A. Tanatar, T. Ishiguro, H. Tanaka, and H. Kobayashi, *Phys. Rev. B*, **66**, 134503 (2002).
5. P. Fulde and R. A. Ferrell, *Phys. Rev.* **135**, A550 (1964).
6. A. I. Larkin and Yu. N. Ovchinnikov, *Zh. Eksp. Teor. Fiz.* 47, 1136 (1964); or *Sov. Phys. JETP* **20**, 762 (1965).
7. J. M. Williams et al., *Organic Superconductivity*, V. Z. Kresin and W. A. Little, eds. Plenum, NY (1990).
8. T. Ishiguro, K. Yamaji, and G. Saito, Organic Superconductors, 2nd ed. Heidelberg, Springer (1998).
9. V. Z. Kresin, H. Morawitz, and S. A. Wolf, *Mechanisms of Conventional and High T_c Superconductivity,* University Press, Oxford (1993).
10. J. Caulfield, W. Lubczynski, F. L. Pratt, J. Singleton, DYK Ko, W. Hayes, M. Kurmoo, and P. Day, *J. Phys.: Condens. Matter* **6**, 2911 (1994).
11. T. Ishiguro, *J. Phys. IV France* **10**, 186 (2000).
12. A. M. Clogston, *Phys. Rev. Lett.* **9**, 266 (1962).
13. B. S. Chandrasekhar, *Appl. Phys. Lett*. **1**, 7 (1962).
14. K. Maki, *Phys. Rev.* **148**, 362, (1966).
15. C. Martin, C. C. Agosta, S. W. Tozer, H. A. Radovan, E. C. Palm, T. P. Murphy, and J. L. Sarrao, *Phys. Rev. B* **71** 020503 (2005). A more detailed discussing of calculating the Pauli limit can be found at http://physics.clarku.edu/superconductor/PauliFold/CalculatingHP.html.
16. F. Zuo, J. S. Brooks, R. H. McKenzie, J. A. Schlueter, and J. M. Williams, *Phys. Rev. B* **61**, 750 (2000).
17. Z. Bayindir, Ph.D thesis, Clark University, 2002
18. R. A. Klemm, A. Luther, and M. R. Beasley, *Phys. Rev. B* **12**, 877 (1975).
19. J. Singleton, *Rep. Prog. Phys.* **63**, 1111 (2000) or J. Singleton, P. A. Goddard, A. Ardavan, N. Harrison, S. J. Blundell, J. A. Schlueter, and A. M. Kini, *Phys. Rev. Lett.* **88**, 037001 (2002).
20. C. C. Agosta, T. Coffey, Z. Bayindir, I. Mihut, C. Martin, and M. Tokumoto, *Physical Phenomena in High Magnetic Fields IV*, G. Boebinger, Z. Fisk, L. P. GorKov, A. Lacerda, J. R. Schrieffer, eds., World Scientific, 333, (2002).

21. Y. Shimojo, S. Kamiya, M. A. Tanatar, E. Ohmichi, A. E. Kovalev, T. Ishiguro, H. Yamochi, G. Saito, J. Yamada, H. Anzai, N. D. Kushch, R. B. Lyubovskii, R. N. Lyubovskaya, and E. B. Yabugskii, *Synth. Met.* **133–134**, 197 (2003).
22. P. A. Mansky, P. M. Chaikin, and R. C. Haddon, *Phys. Rev. B* **50**, 15929 (1994).
23. Z. Bayindir, C. Martin, I. Mihut, L. DeViveiros, T. Coffey, C. C. Agosta, and M. Tokumoto, *Synth. Met.* **120**, 723 (2001).
24. M. Tinkham, *Introduction to Superconductivity* McGraw-Hill, NY (1985).
25. S. W. Tozer, To be published.
26. T. Coffey, Z. Bayindir, J. F. DeCarolis, M. Bennett, G. Esper, and C. C. Agosta, *Rev. Sci. Instrum.* **71**, 4600 (2000).
27. This is a modification of the gasket described in R. Boehler, M. Ross, D. B. Boercker, *Phys. Rev. Lett.* **78**, 4589 (1997).
28. R. A. Forman, G. J. Piermarini, J. D. Barnett, and S. Block, *Science* **176**, 284 (1972).
29. A.-K. Klehe, T. Biggs, C. A. Kuntscher, A. M. Kini, and J. A. Schlueter, cond-mat/0405445
30. J. S. Brooks, X. Chen, S. J. Klepper, S. Valfells, G. J. Athas, Y. Tanaka, T. Kinoshita, N. Kinoshita, M. Tokumoto, H. Anzai, and C. C. Agosta, *Phys. Rev. B* **52**, 14457 (1995).
31. We are using our unpublished results measured down to 50 mK here, as they were measured using the same TDO technique.
32. P. M. Tedrow and R. Meservey, *Phys. Rev. B* **8**, 5098 (1973).
33. T. Schneider and A. Schmidt, *Phys. Rev. B* **47**, 5915 (1993).
34. L. N. Bulaevskii, *Int. J. Mod. Phys. B* **4**, 1849 (1990).
35. S. A. Ivanov, C. C. Agosta, T. Coffey, B. W. Fravel, and K. Montgomery, *Physical Phenomena in High Magnetic Fields III*, Z. Fisk, L. P. GorKov, J. R. Schrieffer, eds., World Scientific, 241 (1999).