Pairing symmetry and two-fluid behavior in the heavy fermion superconductor $CeCoIn_5$

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CeCoIn₅ has recently been the subject of much controversy regarding the symmetry of its order parameter as well as the possibility of multi-band superconductivity. Experiments have been done which show *d*-wave pairing as well as evidence for two-fluid-like behavior. In this paper, I will first briefly review what is know about heavy fermion superconductors. I will then discuss two majors experiments which have been done recently on CeCoIn₅. I will finish by discussing some of the theory explaining the two-fluid model and what could be done to resolve the dispute.

I. INTRODUCTION

Heavy fermion compounds first gained the interest of condensed matter physicists in the 1970s. These compounds are characterized by anomalously large values of the linear contribution to the specific heat (290 mJ/mol K^2 in the case of CeCoIn₅), corresponding to large effective mass charge carriers. Heavy fermions also tend to exhibit very unusual phenomena, such as proximity to magnetic instabilities, quantum critical points, non-Fermi liquid behavior, and unconventional superconductivity. Superconductivity was first discovered in heavy fermions in CeCu₂Si₂. This discovery sparked much interest since these systems show highly non-BCS behavior. CeCoIn₅ has the highest heavy fermion T_c to date at 2.3K and has a highly unusual pairing symmetry. The crystal structure of $CeCoIn_5$ has alternating layers of CeIn₃ and CoIn₂, giving it a quasi-2D structure and some similarity to the high- T_c materials. The superconductivity in CeCoIn₅ seems to arise from magnetic interactions, leading to an anisotropic pairing potential which in turn can have line nodes in the order parameter. $CeCoIn_5$ also has a very complicated Fermi surface, which is suspected to lead to two-fluid-like behavior in which there are two types of electrons: 'heavy' electrons, which condense at T_c , and 'light' electrons, which do not. In this paper, I will first give a brief description of heavy fermion superconductivity. Then I will describe the experiments which led to the expectation of two-fluid behavior in $CeCoIn_5$. I will finish by discussing some of the theory specific to the two-fluid model and further experiments which could be done.

II. GENERAL THEORY OF HEAVY FERMION SUPERCONDUCTIVITY

As stated above, heavy fermion materials contain elements whose f-shell electrons are strongly correlated, thus giving rise to a large effective mass in the quasiparticle excitations. Due to the large Coulomb repulsion between the electrons and their strongly correlated behavior, it is expected that the pairs formed in the heavy fermion superconducting state will not be s-wave. Instead, they are expected to pair up in the asymmetric p-wave or the anisotropic d-wave schemes in order to avoid the large spatial overlap associated with the symmetric s-wave state. The first observation of an anisotropic pairing occurred when Ott et al. [1] observed a T^3 power law in the specific heat of UBe₁₃ as opposed to the exponential behavior expected in a typical BCS superconductor. This type of pairing is similar to what is seen in ³He and, indeed, the analogy between the two-fluid model in ³He and the heavy fermion superconductors is quite useful [2]. It is thought that instead of having a phonon-mediated superconducting state, the heavy fermion superconducting state is mediated by exchange of antiferromagnetic spin fluctuations. This belief is reinforced by the fact that many heavy fermion systems are in or near an antiferromagnetic state. This type of interaction would lead to a $d_{x^2-y^2}$ pairing. The gap equation for this anisotropic state is $\Delta(k) = \Delta_0 [\cos(k_x a) - \cos(k_y a)]$. It is easy to see that the gap will be zero when $k_x a = n\pi/2$ and $k_y a = m\pi/2$ or when $k_x a = n\pi$ and $k_y a = m\pi$ where m and n are both even or odd integers. This corresponds to line nodes along the (110) direction of the crystal. The nodes in the gap cause an excess of quasiparticles along those lines, which can be seen using tunneling measurements. By studying the nodal structure of the superconductor, as well as doing other measurements, one can determine what type of pairing symmetry the material exhibits.

III. NONMAGNETIC IMPURITY DOPING

In 2005, Tanatar et al. [3] performed an experiment in which they doped $CeCoIn_5$ with Lanthanum as a nonmagnetic impurity. Since magnetic fluctuations are thought to be responsible for superconductivity in many heavy fermion materials, nonmagnetic impurities act as pair breakers and create quasiparticles. These quasiparticles can be analyzed by studying the specific heat (C) and thermal conductivity (κ) as a function of doping level. For a d-wave superconductor with line nodes, it is expected that C/T will vary linearly with doping level while κ/T should be a constant. This can be understood by applying a simple kinetic theory to the nodal quasiparticles. $\kappa_{0S}/T \propto \gamma_{0S} v_F^2/\Gamma$ where v_F^2 is the Fermi velocity of the quasiparticle and Γ is the scattering rate which is proportional to the impurity concentration, x [3]. It is expected that $\gamma_{0S} \propto x$ since the density of states for the superconductor varies linearly with energy. This type of linear dependence is indeed borne out by the data. Inserting the linear dependence of the specific heat coefficient on x into the equation for the thermal conductivity, we see that $\kappa_{0S}/T \propto x v_F^2/x = v_F^2$ thus implying that κ_{0S} should be a constant function of x. The data in figure 1 show clearly that the thermal conductivity is not a constant, but varies roughly linearly with x, though the specific heat behaves in the expected way.



FIG. 1: (a) Temperature dependence of specific heat as a function of doping level (x). Inset: residual C/T as a function of x. (b) Temperature dependence of thermal conductivity as a function of x. Inset: residual κ/T as a function of x. (both figures taken from [3])

This discrepancy can be explained using the multiband scenario which I will discuss briefly here and in more detail in a later section. In this scenario, some of the charge carriers condense and participate in superconductivity while the rest do not. The complex Fermi surface of CeCoIn₅ is what makes this possible. The hole band 14 and electron band 15 are quasi-2D and contain the large mass carriers leading to the heavy fermion behavior, while the hole band 13 and electron band 16 form a three-dimensional surface with much lighter mass carriers. By assuming that the lighter carriers to not participate in superconductivity, the apparent discrepancies in the data can be explained. The lighter mass carriers will have a higher velocity and therefore will be much more important for thermal conductivity than specific heat; thus their effects will be seen more strongly in the measurement of κ . Indeed, by assuming that some fraction, $\eta=0.16$ of the carriers are uncondensed and have the same temperature dependence as the normal state while the condensed carriers have some constant value, the data can be fit quite well (see figure 2). In that figure, the line



FIG. 2: Residual κ/T for the superconducting (circles) and normal (triangles) states as a function of the normal-state residual resistivity. The dashed line is the constant value for κ/T of the condensed carriers while the dotted line is the dependence of the uncondensed carriers. For $\eta=0.16$ and $\kappa_{0node}/T=1.4$ mW cm⁻¹K⁻² (where κ_{0node} is the constant value of the superconducting contribution to the thermal conductivity), the data is fit quite well. (figure from [3])

nodes in the superconducting state were assumed to have a constant value of $\kappa_{0node}=1.4$ mW/cm K². The rest of the data was then fit by assuming that $\kappa/T = \eta \kappa_{0N}/T + \kappa_{0node}/T$. The multiband theory will be expanded upon more in a later section.

IV. POINT CONTACT SPECTROSCOPY MEASUREMENTS

Point contact spectroscopy (PCS) measurements on $CeCoIn_5$ have been done by multiple groups in the hopes of determining conclusively the order parameter symmetry, location of line nodes, and the possibility of multiple bands [4–8]. In PCS, a small, conductive probe (usually made from a normal metal like Au) is placed in contact with the sample, thus creating a type of Josephson junction. A voltage is then applied across this contact and the differential conductance, dI/dV, is measured. There are generally two types of processes by which particles can get across the junction. The first is Andreev reflection, in which an electron from the normal metal is transmitted into the superconductor and a hole is retroreflected. This leads to a current which is twice what it would be when the superconductor is in the normal state. The second process by which electrons can cross the junction is through quasiparticle tunneling. In this process, electrons from the normal metal tunnel into the quasiparticle states in the superconductor. This only happens in nontransparent junctions. Since Andreev reflection only occurs when the bias voltage is less than the superconducting gap, outside the gap the current returns to its usual value, thus providing information about the gap magnitude. By performing PCS at various directions on the sample, dependence of the gap on crystal direction can be mapped out and the pairing symmetry deduced. The Blonder-Tinkham-Klapwijk (BTK) theory was developed in order to predict the shape of the dI/dV curves based on these two types of tunneling. This theory was derived for s-wave superconductors but has been extended to handle d-wave superconductors. It is based on a dimensionless parameter, $Z_{eff} = \sqrt{Z_0^2 + (1-r)^2/4r}$ where Z_0 is due to the physical barrier and r is the ratio of the Fermi velocity in the normal metal to that in the superconductor.

Although PCS has a number of advantages, it also has some major drawbacks. In par-

ticular, if the contact made with the sample is too large, the experiment is no longer in the ballistic (Sharvin) regime and many anomalous results can be seen. A contact is considered to be in the Sharvin regime if the dimensions of the contact are smaller than the electronic scales of the material [5]. This means that the contact radius, a, must be much smaller than the electronic mean free path, l. However, determining if the experiment is in the Sharvin regime is not easy, as the radius of the contact is generally not known precisely and the mean free path of a material can vary from sample to sample. For this reason, the PCS results on CeCoIn₅ have varied wildly depending on the group. Only lately has some type of consensus emerged. In addition to this difficulty, interpreting PCS results for d-wave superconductors is complex because the position of the maximum in the dI/dV spectrum does not only depend on the gap, but also the order parameter symmetry, the barrier strength, and other microscopic considerations. Since d-wave superconductors can break time-reversal or point-group symmetry, this adds to the difficulty.

Although many groups have done PCS experiments, I will focus on two papers whose results have been interpreted in terms of multiple band superconductivity. The data published by Goll et al. [5], Park et al. [6], Greene et al. [7] is similar to the recent data published on the arXiv [8]. In addition to the arXiv paper, I will also briefly discuss the paper by Rourke et al. [4] which shows quite different data but reaches a similar conclusion. It should be noted that at least two groups believe that the Rourke data is not in the ballistic regime and thus that the data is anomalous [9–11]. This controversy will not be further discussed here.

The most recent work by Park et al. [8] shows more convincing evidence for a two-fluid type behavior in CeCoIn₅ and $d_{x^2-y^2}$ pairing. In this paper, PCS measurements were done on three faces of $CeCoIn_5$: (001), (110), and (100). Over two hundred junctions were measured, thus suggesting that the features seen are intrinsic to the material rather than aberrations. The features along the (001) and (100) are qualitatively the same, while the features along the (110) direction are quite different. This is consistent with the theory that $CeCoIn_5$ is a *d*-wave superconductor, as the nodes lie along the (110) direction. In figure 3, these disparities can be seen, with the (100) data showing a rather flat structure near zero bias while the (110) data show a peaked structure at zero bias. A key signature of a d-wave node is the shape of the zero-bias conduction peak (ZBP). In these systems, the ZBP arises from surface states bound by interference between the phases of consecutively Andreev-reflected quasiparticles. This will cause a peaked structure along nodal directions while a hump structure associated with Andreev bulk states is seen along other directions. It should be noted that the value of the ZBP in this data is much smaller than the theoretical prediction. In figures 3 (c) and (d), the calculated BTK curves are shown for antinodal and nodal junctions, respectively. For a particular value of Z_{eff} , the antinodal simulation can reproduce the flat behavior seen in the (100) direction, whereas for the nodal junction, a peaked structure is seen regardless of the value of Z_{eff} . Park et al. [8] conclude that since the flat conductance shape can occur only for an antinodal junction with $Z_{eff} \sim 0.28$, the (100) direction must be antinodal. They also state that since the peaked shape of the (110)direction cannot happen in an antinodal junction except for small Z_{eff} and such a small Z_{eff} is exceedingly unlikely, the (110) direction must be the nodal direction. Their data certainly support the idea of $d_{x^2-y^2}$ pairing, but does not definitively rule out other pairing mechanisms.

The spectroscopic data also support the two-fluid interpretation. If it is assumed that there is a fluid of uncondensed electrons coexisting with a fluid of superconducting electrons,



FIG. 3: Normalized conductance spectra of CeCoIn₅/Au junctions along the (a) (100) direction and (b) along the (110) direction. The data have been shifted vertically. Calculated conductance curves using the BTK model for *d*-wave superconductors with $\Gamma = 0$ and T = 0 for (c) antinodal and (d) nodal junctions. Figures taken from [8].



FIG. 4: Fit to data using the modified BTK model. Parameters for the curve are $\omega_h = 0.51$, $\Delta = 066 \mu \text{eV}, \Gamma = 95 \mu \text{eV}, Z_{eff} = 0.28, \eta = 1, \Lambda = 5 \text{ meV}$ and $\epsilon_0 = -2.1 \text{ meV}$. Figures taken from [8].

the BTK theory must be modified in the following way: $\frac{dI}{dV}(V) = \omega_h \frac{dI}{dV}|_h(V) + (1-\omega_h)\frac{dI}{dV}|_l(V)$ [8]. In essence, it is the sum of two parallel conducting channels, where ω_h is the weighting factor related to the heavy fermion spectral weight. When fitting the data, a Lorentzian density of states was assumed with η as the peak height, Λ as the half-width, and ϵ_0 as the center of the curve. As can be seen in figure 4, the fit to data is quite good.

Fit parameters are given in the figure caption, where Δ is the energy gap, Γ is the quasiparticle lifetime broadening factor, and the other parameters have been defined. The fit gives $2\Delta/k_BT_c = 6.05$, which is consistent with strong coupling. Although the fit is very good, it should be noted that there are many fit parameters and equally good fits have been obtained with larger values of ω_h , though this does lead to smaller Δ and larger Γ values, which leads to an unphysical temperature dependence of Γ .

Rourke et al. [4] also did PCS experiments on $CeCoIn_5$ and, though their data looks qual-



FIG. 5: Temperature dependence of the two gap amplitudes, Δ_1 and Δ_2 . At zero temperature, the gaps approach the values $\Delta_1 = 0.95 \pm 0.15$ meV and $\Delta_2 = 2.4 \pm 0.3$ meV. (figure from [4])

itatively different, they came to similar conclusions. As stated earlier, some groups believe that this data is not real, as the contact may not have been in the ballistic regime. Their data was fit well by using the *d*-wave BTK formalism with two parallel or serial channels depending on the junction impedance. This was explained by assuming two different gaps. When the bulk spectra from two different order parameters coexist, they add in parallel, while when both bulk and surface states exist, they add in series. This interpretation is similar to that used by Tanatar et al. [3], Park et al. [6], but not quite. This paper assumes two gaps of differing magnitude, while the others assume one gapped fluid and one nongapped fluid. In figure 5, the temperature evolution of the two gaps can be seen.

Before closing this section, I would like to restate that, although the PCS measurements provide evidence for multiple bands, they do not definitively prove it. PCS experiments are difficult to do in the best conditions, and *d*-wave superconductors provide even more difficulty.

V. OTHER EXPERIMENTS

Many other experiments have been done on CeCoIn₅. Here I will discuss a few which could be explained by an application of the two-fluid model. Bel et al. [12] measured the Nernst and Seebeck coefficients of CeCoIn₅ and found an anomalously large sublinear Nernst signal below 18K. The Nernst effect is observed when a magnetic field and temperature gradient are applied normal to each other to a sample. An electric field will be induced in the remaining direction. The Nernst coefficient is defined as $N_i = \frac{E_i/B}{\nabla T}$ where *i* is a direction. Bel et al. [12] observed a very large signal in the longitudinal and the transverse directions. There are two rather striking features in their data: the electric field becomes more parallel to the thermal current with increasing magnetic field and as the temperature goes to zero in the zero-field limit, the electric field produced by a longitudinal heat current becomes purely transverse. It is thought that by applying the multiband scenario to the Nernst effect, these unusual behaviors could be explained.

Xiao et al. [13] did angular-dependent torque measurements on CeCoIn₅. They found that $\gamma \equiv \sqrt{m_c^*/m_a^*}$ where m_i^* is the cyclotron mass in the *i*th direction was not a constant, as expected, but was field and temperature dependent. This could be explained by two gaps which have different temperature and field evolutions. These observations are similar to those seen in the high-T_c materials, again lending credence to the idea that, by understanding these materials, we can better understand the high- T_c superconductors.

VI. THEORY

In general, it is thought that the two-fluid behavior in $CeCoIn_5$ is caused by the shape of the Fermi surface, which consists of both two-dimensional sheets containing the heavy electrons and three dimensional pockets of light electrons. The heavy electrons on the 2D sheets condense at the transition temperature while the light electrons either do not condense or have a very small gap. The theoretical shape of some of the relevant Fermi surfaces can be see in figure 6.



FIG. 6: Hole band 14, electron band 15, hole band 13, and electron band 16. The first two bands are quasi-two-dimensional, contain heavy electrons, and cause superconductivity while the latter two are three dimensional and contain light electrons which do not condense. Figures are band structures calculated by Maehira et al. [14]. These calculations agree well with experiment, saving that band 16 has not yet been observed experimentally [15, 16].

Prior to the work done on PCS and thermal conductivity/specific heat, Nakatsuji et al. [17] did a calculation in which they examined the two-fluid description of CeCoIn₅. They found that there are indeed two fluids, a coherent heavy fermion state and a background lattice of noninteracting Kondo impurity centers. CeCoIn₅ has three energy scales: the single ion Kondo temperature $T_K=1.7$ K, the site-to-site coupling energy $T^*=45$ K, and the crystal electric field splitting $T_{CEF}=120$ K. When CeCoIn₅ is diluted with La on the Ce site, only T^* changes, making this a good way to separate the Kondo behavior from the heavy fermion behavior. Below T^* , the emergence of the coherent heavy fermion state can be seen and its fraction, f, can be determined as a function of temperature. It turns out that f increases linearly with decreasing temperature and ultimately saturates to 0.9. This form for f was determined by looking at the magnetic f-electron part of the specific heat, C_{MAG} .

 C_{MAG} can be decomposed into C_{KI} , the single ion contribution of the f electrons and C_{HF} , the heavy fermion fluid contribution, in the following way: $C_{MAG}/T = [1 - f(T)](C_{KI}/T) + f(T)(C_{HF}/T)$. C_{MAG} is determined by subtracting the lattice specific heat (that of LaCoIn₅) from the specific heat of the Ce_{1-x}La_xCoIn₅. By plotting C_{MAG} as a function of C_{KI} and determining the slope of the line, f can be found as a function of x. C_{KI} was determined from the specific heat of the Ce-La alloys at low Ce concentrations. As can be seen in figure 7, for concentrations between 0 and 0.25, the slope is saturated at 0.1 with an offset of 290 mJ/mol-Ce K² which is just the linear contribution to the specific heat (i.e. the effective mass of the electrons). This indicates that the magnetic specific heat at low temperature and impurity concentration is just a linear function of the Kondo specific heat. Thus it



FIG. 7: The linear part of the magnetic contribution to specific heat versus the linear part of the single impurity limit. The x values given in the figure are the impurity (La) concentration from top to bottom. Figure from [17].

can be concluded that for low temperatures, the ground state has two fluids: a 10% Kondo impurity fluid (the single-ion contribution) and a 90% heavy-fermion fluid (the coherent contribution due to intersite coupling). This can be understood by saying that each felectron acts as though it is 10% Kondo impurity and 90% heavy fermion. Nakatsuji et al. [17] also calculated the Wilson ratio (the dimensionless ratio of the low temperature spin susceptibility to the specific heat coefficient), $R_W = \alpha \chi/(C/T)$ where $\alpha = (\pi^2 k_B^2/3\mu_B^2)$, for the heavy fermion fluid and found that it is 2.0, the result expected for the Kondo impurity case.

While these results are very interesting, what we would like to determine is the temperature dependence of f for pure CeCoIn₅. This is difficult, as C_{HF}/T , C_{KI}/T , C_{MAG}/T , and f are all temperature dependent. Fortunately, the dependence of C_{KI}/T and C_{MAG}/T are already known experimentally. To determine the dependence of the other quantities, we should first look at the magnetic susceptibility, writing it similarly to C_{MAG}/T : $\chi(T) = [1 - f(T)](\chi_{KI}/T) + f(T)(\chi_{HF}/T)$. We then assume that below T^* , the Wilson ratio (defined above) of the heavy fermion component is always 2: $\chi_{HF}(T) = 2C_{HF}(T)/\alpha T$. There are now three equations and three unknowns ($\chi_{HF}(T)$, $C_{HF}(T)$, and f(T)), we can combine them and determine the temperature dependence from experiment. As can be seen in figure 8, f(T), which plays the role of order parameter, increases linearly with decreasing temperature below T^* , saturating to 0.9 at near 2K. This behavior is qualitatively similar to the increase in fraction of superfluid with decreasing temperature seen in ³He.

In their paper, Nakatsuji et al. [17] give two quite convincing checks of their analysis. First, they assume that the magnetic part of the electrical resistivity comes from scattering off the Kondo impurity centers only. This implies that the magnetic part of the resistivity (defined as the difference in resistivity between CeCoIn₅ and LaCoIn₅) should be the same as the Kondo impurity resistivity multiplied by (1-f). The Kondo impurity resistivity is determined by taking the measured Ce single impurity resistivity in LaCoIn₅ and scaling to 100% Ce. As can be seen in figure 9, the data is fit remarkably well.

Secondly, the magnetization, M, as a function of applied field, H can be calculated. In a similar vein as previously, M can be written as $M = f M_{HF} + (1-f)M_{KI}$. The heavy fermion component should just give a linear response $(M_{HF} = \chi_{HF}H)$, while the local moment part



FIG. 8: The relative fraction of the heavy fermion fluid, f(T), as a function of temperature. Dashed line is a linear fit. Figure from [17].



FIG. 9: Magnetic part of resistivity, defined as the difference between $CeCoIn_5$ and $LaCoIn_5$. Red open symbols are $CeCoIn_5$ and blue open symbols are the magnetic resistivity measured in the single impurity limit. Black line is resistivity calculated by multiplying the single impurity limit data by (1-f). Figure from [17].

will be a sum of a van Vleck linear part and a Brillouin-type nonlinear response due to saturation of local moments $(M_{KI} = g\mu_B \{2a(g\mu_B H) + b \tanh [bg\mu_B H/k_B(T + T_K)]\}$, where a and b are constants from the CEF splitting). The field dependence of M can then be calculated with no fitting parameters. The result is plotted in the inset to figure 9. The data is reproduced well above the upper critical field of 5T. These checks lend further evidence to the two-fluid model.

VII. FURTHER WORK

A problem with the measurements which have been done so far is that, while supporting the $d_{x^2-y^2}$ symmetry proposition, they do not rule out other symmetries. One way to unambiguously determine the pairing symmetry of these compounds would be to do phasesensitive measurements. Since for a *d*-wave pairing symmetry, the order parameter changes sign with different *k*-space directions (see figure 10a), a phase-sensitive measurement would be able to distinguish this. In this type of experiment, Josephson junctions are made at the corners, opposite sides, or same sides of the crystal. We will consider only the corner junction here, to illustrate the concept. For further details, see the review article by Van Harlingen [18]. For the corner method, a Josephson junction is created between one side of the crystal and a conventional superconductor. A second junction is created on a perpendicular side and the two are connected by the same conventional superconductor, thus creating a loop (see figure 10b). A magnetic field is then applied perpendicular to the loop. The Josephson relation tells us that $I = I_{ca} \sin \phi_a + I_{cb} \sin \phi_b$ where I_{ci} is the critical current of the *i*th junction and ϕ_i is the phase across the *i*th junction. By mapping out the change in current as a function of applied field, the difference in phase between the two Josephson junctions can be determined. By repeating this experiment with junctions at various parts of the crystal, the entire order parameter phase can be mapped out and the pairing symmetry determined conclusively.



FIG. 10: Sketch of the $d_{x^2-y^2}$ order parameter in a crystal. Drawing of a corner squid interferometer. Figures taken from [18].

VIII. CONCLUSION

Heavy fermion compounds and CeCoIn₅ in particular exhibit unique and interesting properties like unconventional superconductivity. In the case of CeCoIn₅, it is thought that the pairing interaction is mediated by exchange of antiferromagnetic spin fluctuations which in turn causes an unusual pairing symmetry. Among the many experiments done on this compound, PCS and measurements of the temperature dependence of the thermal conductivity and specific heat have been among the most helpful in understanding its properties. It has been found that CeCoIn₅ pairs up in a *d*-wave state, probably the $d_{x^2-y^2}$ state. CeCoIn₅ has also exhibited behavior consistent with it having two fluids: heavy, condensed electrons and light, uncondensed electrons. This is caused by the complex nature of its Fermi surface, with the heavy electrons residing on the two-dimensional sheets and the light electrons in the three dimensional pockets. Since CeCoIn₅ has a structure similar to that of the high- T_c compounds, it can be hoped that greater understanding of this compound can lend insight to this yet unsolved problem.

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