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### Authors

Park, WK  
Sarao, JL  
Thompson, JD  
[et al.](#)

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# Andreev reflection spectroscopy of the pure and Cd-doped heavy-fermion superconductor CeCoIn<sub>5</sub>: Detecting order parameter symmetry and competing phases

W.K. Park<sup>a,\*</sup>, J.L. Sarrao<sup>b</sup>, J.D. Thompson<sup>b</sup>, L.D. Pham<sup>c</sup>, Z. Fisk<sup>d</sup>, L.H. Greene<sup>a</sup>

<sup>a</sup>Department of Physics, University of Illinois at Urbana-Champaign, Urbana, IL 61801, USA

<sup>b</sup>Los Alamos National Laboratory, Los Alamos, NM 87545, USA

<sup>c</sup>University of California, Davis, CA 95616, USA

<sup>d</sup>University of California, Irvine, CA 92697, USA

## Abstract

Andreev reflection conductance spectra are obtained on nanoscale ballistic junctions between Au tips and single crystals of the pure and Cd-doped heavy-fermion superconductor CeCoIn<sub>5</sub>. Background conductance asymmetry starting at the heavy-fermion coherence temperature  $T^*$  ( $\sim 45$  K) and increasing with decreasing temperature down to  $T_c$  (2.3 K) signifies the emerging heavy-fermion liquid in CeCoIn<sub>5</sub>. Below  $T_c$ , enhancement of the sub-gap conductance arises from Andreev reflection, but the Blonder–Tinkham–Klapwijk theory dictates that the Fermi velocity mismatch should yield no Andreev reflection. The signal we do observe is several times weaker than that observed in conventional superconductors, but consistent with other heavy-fermion superconductor data reported. Data taken in the (001), (110), and (100) orientations provide consistent and reliable spectroscopic evidence for  $d_{x^2-y^2}$  symmetry of the superconducting order parameter. Conductance spectra on the (100) surface of 10% Cd-doped CeCoIn<sub>5</sub> show intriguing behaviors following antiferromagnetic and subsequent superconducting transitions.

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**Keywords:** Heavy-fermion superconductor; Andreev reflection; d-wave symmetry; Competing phases

## 1. Introduction

The so-called 1-1-5 family of heavy-fermion compounds continue to draw much attention because of their unique physical properties. Although it has been well established that the superconducting order parameter of CeCoIn<sub>5</sub> has line nodes consistent with d-wave symmetry [1], its detailed structure, e.g., location of nodes, remains to be clarified [2–4]. We have reported Andreev reflection [5] spectroscopy results on CeCoIn<sub>5</sub> along three different crystallographic orientations, providing the first spectroscopic evidence for  $d_{x^2-y^2}$  symmetry [6–8]. However, quantitative understanding of the reduced Andreev conductance, which has been commonly observed in most heavy-fermion superconductors [6,9,10], is still lacking. Here, we propose to modify the

Blonder–Tinkham–Klapwijk (BTK) model [11,12] by considering energy-dependent density of states and two-fluid behavior [13] in heavy fermions.

Recently, Pham et al. [14] have reported intriguing phase diagrams in Cd-doped 1-1-5 compounds. These chemically tuned electronic systems provide unique avenues for the study of rich phenomena including competing/coexisting phases, quantum criticality, etc. Our conductance spectra on 10% Cd-doped CeCoIn<sub>5</sub> reveal signatures for coexisting/competing phases of superconductivity and antiferromagnetism, in line with the reported phase diagram [14].

## 2. Experiments

All samples studied in this work are single crystalline. For Andreev reflection measurements along different crystallographic orientations, single crystal surfaces along three major axes  $\{(001), (110), \text{and } (100)\}$  are prepared

\*Corresponding author. Tel.: +1 217 265 5010; fax: +1 217 244 8544.

E-mail address: [wkpark@uiuc.edu](mailto:wkpark@uiuc.edu) (W.K. Park).

by embedding into epoxy, cutting, and polishing. Crystallographic orientations are confirmed by X-ray diffraction analysis [8]. Electrochemically polished Au tips are used as counter-electrodes. Point-contact junctions between single crystal samples and Au tips are formed using our home-built Cantilever-Andreev-Tunneling rig [15]. Differential conductance data are taken as a function of temperature and magnetic field using standard lock-in techniques. Here, positive voltage means that the CeCoIn<sub>5</sub> electrode is biased positively.

### 3. Results and discussion

Normalized conductance spectra of CeCoIn<sub>5</sub> along the two in-plane directions are displayed in Fig. 1, together with calculated conductance curves using the d-wave BTK model [12]. Obtained data reveal strikingly different characteristics at the lowest temperature, namely, rather flat shape for the (100) junction vs. sharp cusp-like feature for the (110) junction. The difference in the characteristics (rounded vs. triangular) is also visible even at higher temperatures despite an increased thermal smearing.

According to the d-wave BTK model [12], conductance behavior as a function of the barrier strength,  $Z$ , is dramatically different along different directions, as demonstrated in Figs. 1(c and d). For an anti-nodal junction, the zero-bias conductance is reduced with increasing  $Z$ , thus, developing a flattened region for  $Z \sim 0.25$  and a double-peak structure for larger  $Z$ . In contrast, a nodal junction exhibits sharper and sharper features with increasing  $Z$ . It shows a conductance peak at zero-bias in the tunneling limit, which originates from the zero-energy Andreev

bound states (ABS) formed on symmetry-breaking surfaces. Broader features for smaller  $Z$  are essentially due to the smearing of ABS to finite energy caused by the higher junction transparency. In practical junctions,  $Z$  is always non-zero because of either dielectric layer or Fermi velocity mismatch [11]. This is likely the case for Au/CeCoIn<sub>5</sub> due to the disparate Fermi velocities. Then, it is reasonable to identify the (100) junction as along the anti-nodal direction, whereas the (110) junction as along the nodal direction. Therefore, our conductance spectra are consistent with  $d_{x^2-y^2}$  symmetry [2,4] rather than  $d_{xy}$  symmetry [3].

As we already reported [6,8,9], our conductance spectra cannot be explained quantitatively by the BTK theory and its modified versions. Seeking a clue to this problem, here we note two experimental observations on CeCoIn<sub>5</sub>: the two-fluid behavior reported by Nakatsuji et al. [13] and the existence of unpaired light electrons below the  $T_c$  of CeCoIn<sub>5</sub> reported by Tanatar et al. [16]. The growing spectral weight of the heavy-electron liquid [13] implies that the density of states has stronger energy dependence as the temperature is lowered. One possible origin for such energy dependence is the lattice version of Kondo resonance, whose detailed features in CeCoIn<sub>5</sub> remain to be studied by photoemission, tunneling, etc. Since the point-contact conductance is affected by the electronic density of states of the electrodes [10], it is expected to develop an asymmetry that is enhanced with decreasing temperature. This is exactly what we observe [6,9].

Taking these two experimental reports into consideration, we propose to modify the BTK model to explain our conductance data quantitatively. Here, two parallel

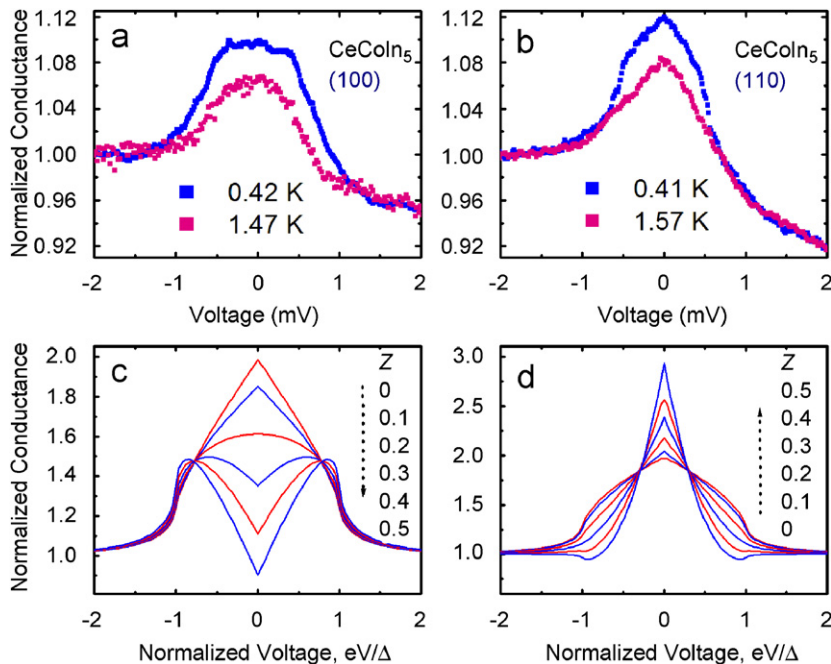


Fig. 1. (color online) (a, b) Normalized conductance spectra of CeCoIn<sub>5</sub> on (100) and (110) surfaces, respectively. (c, d) Calculated conductance curves for anti-nodal and nodal junctions of a d-wave superconductor, respectively.  $Z$  is the barrier strength parameter.

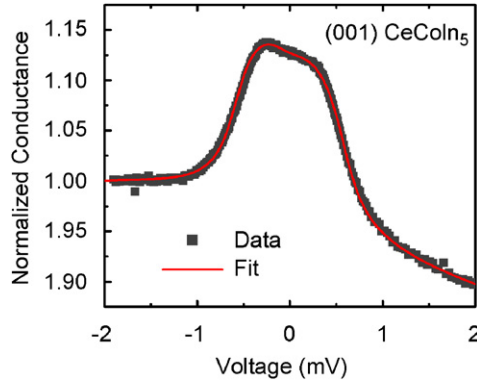


Fig. 2. (color online) Conductance data for (001) CeCoIn<sub>5</sub> taken at 400 mK and the best-fit curve using our modified BTK model.  $G(V) = \omega_h G_h(V) + (1 - \omega_h) G_l(V)$ .  $D(\epsilon) = 1 + \sigma \Gamma^2 / [\Gamma^2 + (\epsilon - \epsilon_0)^2]$ . See the text for details.

conductance channels are assumed, one for superconducting heavy electrons and the other for normal conducting light electrons. The unpaired light electrons contribute to a constant conductance ( $G_l$ ) and the superconducting heavy electrons contribute to the usual BTK-like conductance ( $G_h$ ) with an asymmetric background due to an energy-dependent density of states,  $D(\epsilon)$ . Fig. 2 shows the resultant best fit to the conductance data for an Au/(001) CeCoIn<sub>5</sub> junction at 400 mK. Here, a Lorentzian shape is assumed for the peak in the density of states,  $D(\epsilon)$ , of the heavy-fermion liquid. As seen, our data can be fit very well with the fitting parameters:  $\omega_h = 0.51$ ,  $\Delta = 600 \mu\text{eV}$ ,  $\Gamma_{\text{Dynes}} = 95 \mu\text{eV}$ ,  $Z = 0.28$ ,  $\sigma = 1$ ,  $\Gamma = 5 \text{ meV}$ , and  $\epsilon_0 = -2.1 \text{ meV}$ . It is found that the quality of the fit strongly depends on the heavy-fermion weight factor  $\omega_h$ , implying that our modeling based on the two-fluid picture is quite reasonable. The obtained energy gap ( $\Delta$ ), and the quasiparticle lifetime broadening factor ( $\Gamma_{\text{Dynes}}$ ), are larger, and smaller, respectively, than those reported earlier [9], where a single channel conductance is simulated using the d-wave BTK model [12]. Considering the generality of the two-fluid behavior in heavy fermions [17], our modeling might also be applicable to other heavy-fermion superconductors. Refining this model and further analyses of our data are underway.

Cadmium is known to substitute for indium sites in the basal plane of CeCoIn<sub>5</sub>, creating a hole [14]. 10% Cd doping induces coexisting phases with subsequent anti-ferromagnetic and superconducting transitions at  $T_N \sim 2.9 \text{ K}$  and  $T_c \sim 1.3 \text{ K}$ , respectively.

The zero-bias conductance vs. temperature data taken on the (100) surface of a 10% Cd-doped CeCoIn<sub>5</sub> crystal are plotted in Fig. 3. It shows a non-monotonic behavior, in contrast with that observed in undoped CeCoIn<sub>5</sub> [6,9], which exhibits conductance enhancement due to usual Andreev reflection [5]. This indicates that there must be an additional mechanism for conductance enhancement below  $T_N$ . Conductance spectra at several temperatures are also plotted in Fig. 3. Surprisingly, more complicated behaviors are seen: a broad conductance peak at high

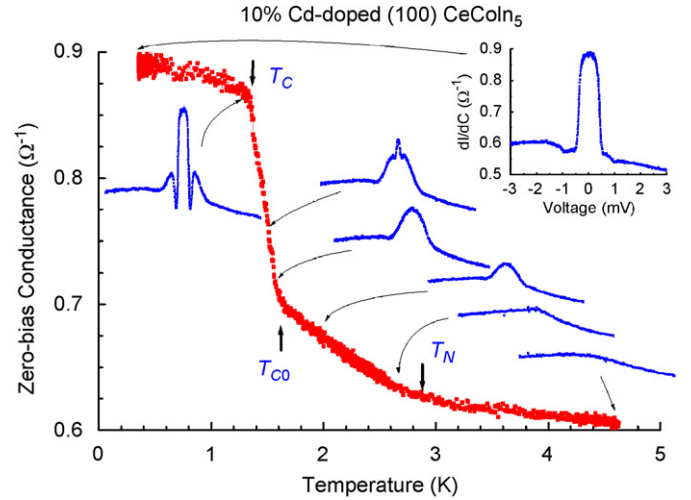


Fig. 3. (color online) Zero-bias conductance of the 10% Cd-doped (100) CeCoIn<sub>5</sub>/Au junction as a function of temperature. Also shown are conductance spectra at several temperatures.

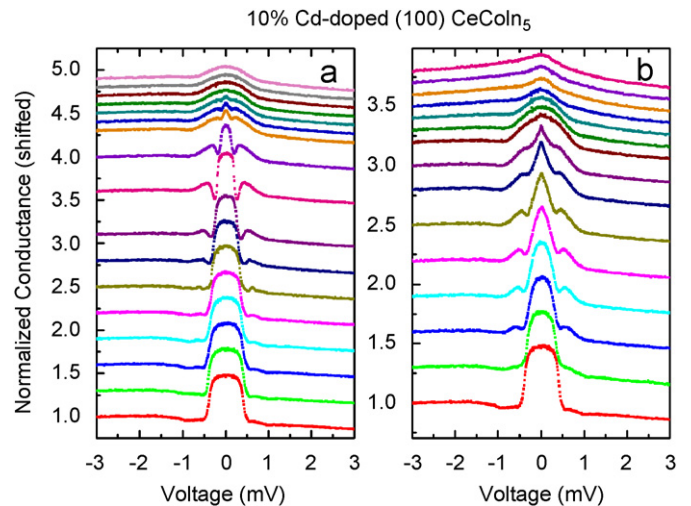


Fig. 4. (color online) Normalized conductance spectra of the 10% Cd-doped (100) CeCoIn<sub>5</sub>/Au junction as a function of (a) temperature and (b) magnetic field at 0.41 K. (a) Temperatures are 0.42, 0.44, 0.55, 0.73, 0.87, 1.01, 1.12, 1.22, 1.32, 1.40, 1.51, 1.54, 1.56, 1.59, 1.68, 1.76, and 1.85 K from the bottom. (b) Magnetic field values are 0, 1, 1.5, 1.75, 2, 2.1, 2.25, 2.5, 3, 4, 5, 6, 7, 8, and 9 T from the bottom.

temperatures and competing conductance channels at lower temperatures.

These complicated features are better seen in the temperature- and field-dependent spectra, as plotted in Fig. 4. The conductance enhancement near zero-bias appears below  $T_N$  with a resultant broad peak over  $\pm 1 \text{ mV}$  range with decreasing temperature down to  $T_{co} \approx 1.6 \text{ K}$ . This temperature corresponds to the zero-resistance temperature of the single crystal, which is a bit higher than the  $T_c$  ( $\sim 1.3 \text{ K}$ ) from specific measurements [14]. Below 1.6 K, there appears an additional conductance feature, a narrower peak centered at zero-bias. With further decrease of temperature, this central peak grows, whereas the

spectral weight of the broad peak reduces. These data are consistent with the existence of two conductance channels competing with each other. This competition persists all the way down to 410 mK, where the signature of the broad peak remains as humps on the shoulders of the central conductance peak. The appearance of the central peak below  $T_{c0}$  indicates it is related to the superconducting phase, e.g., Andreev reflection [5].

Similar behaviors are observed in the magnetic field dependence, as shown in Fig. 4(b). With increasing field along the  $c$ -axis, there appear two competing conductance channels. This competition disappears around  $\sim 3$  T, above which a broad peak similar to the one seen above  $T_{c0}$  is observed. The broad peak is gradually suppressed with increasing field just as with increasing temperature. Therefore, it is likely that the field value of 3 T corresponds to the upper critical field of the superconducting phase.

Although it is quite clear that the central conductance peak is due to superconductivity from both temperature- and field-dependent measurements, the origin of the broad peak is not yet clear. A likely candidate is some kind of bound states. According to Bobkova and coworkers [18], a novel quasiparticle reflection occurs at an interface between an antiferromagnet and a normal-metal or a superconductor. In this configuration, an electron incident from the normal side acquires momentum due to antiferromagnetic order represented by a wave vector  $Q$  and undergoes spin-dependent retro-reflection. If this spin-dependent  $Q$ -reflection is combined with Andreev reflection, low-energy ABS can form on the boundary [18]. Preliminary conductance spectra on the (001) surface of 10% Cd-doped CeCoIn<sub>5</sub> show enhanced-conductance behaviors (either a broad peak or double peaks around zero-bias) but do not show the competing behaviors. Neutron scattering measurements have shown that the antiferromagnetic order in this compound has a wavevector  $Q = (\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$  [19]. This implies that the  $Q$  vectors relevant to the quasiparticle reflection [18] are effectively the same at both (100) and (001) surfaces, thus, explaining the common observation of the broad peak. But what causes the clear difference, namely, observation or non-observation of competing conductance behaviors, is not yet understood. More detailed measurements are underway for samples over a wide range of Cd doping.

#### 4. Conclusion

We report the first spectroscopic evidence for the  $d_{x^2-y^2}$  order parameter symmetry in CeCoIn<sub>5</sub>. A modification of

the BTK model is proposed in order to explain the reduced Andreev signal in normal-metal/heavy-fermion superconductor junctions, taking into account an energy-dependent density of states and the two-fluid behavior in heavy fermions. Conductance spectra on 10% Cd-doped CeCoIn<sub>5</sub> reveal intriguing behaviors as a function of temperature and magnetic field with possible signatures for coexisting/competing phases.

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