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Polar Kerr effect studies of time reversal symmetry breaking states in heavy fermion superconductors

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The connection between chiral superconductivity and topological order has emerged as an active direction in research as more instances of both have been identified in condensed matter systems. With the notable exception of ^{3}He-B, all of the known or suspected chiral – that is to say time-reversal symmetry-breaking (TRSB) – superfluids arise in heavy fermion superconductors, although the vast majority of heavy fermion superconductors preserve time-reversal symmetry. Here we review recent experimental efforts to identify TRSB states in heavy fermion systems via measurement of polar Kerr effect, which is a direct consequence of TRSB.

1. Introduction

The discovery of superconductivity in $CeCu_2Si_2$ in 1979 [1], followed shortly thereafter by UBe_{13} [2] and UPt_3 [3], established the class of heavy fermion superconductors and provided the earliest indications of superconductivity beyond that described by conventional (*s*-wave) BCS theory. Collectively, they are defined by the hybridization of (often magnetic) 4*f* or 5*f* ions with conduction *s*, *p*, or *d* electrons; the resulting quasiparticles have high renormalized masses reflecting the strength of these interactions [4]. To date, this family includes some three dozen compounds that exhibit the greatest variety of superconducting ground states of any family of superconductors, often within complex phase diagrams that also include various types of magnetic ordering and other quantum critical behavior [5].

Of particular interest among the heavy fermion superconductors are those known or suspected to break time-reversal symmetry (TRS). Since the time-reversal operator acts on the orbital component of the order parameter via complex conjugation, the order parameters in TRS-breaking (TRSB) systems have relatively complex coefficients, *e.g.* $\Delta \sim k_x \pm ik_y$, such that $\Delta \neq \Delta^*$. While TRSB superconductivity was first proposed in several early models for UPt_3 and has been most extensively studied in Sr_2RuO_4 [6], TRSB superconductors have in recent years attracted further attention in the context of topological order. The possibility of Majorana and other exotic particles, and incorporation into quantum computation, have now placed these materials at the forefront of condensed matter physics research [7–9].

In unconventional superconductors, a TRSB order parameter may be inferred if the nodal structure is sufficiently well-known. Experimentally, such states are more or less directly observable through μSR, polarized neutron scattering, or detection of circulating edge currents [10–12]. A fourth direct consequence of TRSB, and the focus of this paper, is the appearance of magneto-optical effects [13]. Polar Kerr effect is one such effect that is particularly sensitive to TRSB; its measurement, while challenging, can be especially advantageous in instances where high-quality single crystals are not large enough for bulk neutron scattering experiments, or where complementary techniques yield ambiguous or conflicting results.

This paper reviews our more recent studies of polar Kerr effect (PKE) in heavy fermion superconductors as a step towards identifying the symmetries of their respective order parameters. Section 2 defines PKE and its relationship to TRS and chiral superconductivity. Section 3 describes measurement of PKE in cases where the polarization rotation generated by a TRSB material is nonzero but also extremely weak. Section 4 then reviews the results of such measurements on the foundational heavy fermion superconductors $CeCoIn_5$, UPt_3, and URu_2Si_2. Prior PKE studies of Sr_2RuO_4 [14] are reviewed in [15]. Finally, some future candidates for PKE study are discussed in Section 5.

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2. Polar Kerr effect as a probe of TRSB

Polar Kerr effect (PKE) has been used to study ferromagnetic materials for over a century [16]. However, only since the discovery of unconventional superconductivity has PKE been considered as a possible experimental signature of more exotic types of quantum matter [17]. PKE is described as follows: Consider linearly polarized light (*i.e.* an equal superposition of right- and left-circular polarizations) incident normally upon a material. If the material's complex indices of refraction $\tilde{n}_{+,-}$ for the two circular polarizations differ, the reflected light will be an unequal – in both amplitude and phase – superposition of right- and left-circular polarizations: the reflected polarization state will be elliptical and phase-shifted relative to the incident beam. The Kerr angle θ_K, giving the rotation of the major axis of the reflected beam relative to the incident beam, is then defined by

$$\theta_K = \frac{1}{2}\{\arg[R_{++}] - \arg[R_{--}]\} \tag{1}$$

where R_{++} and R_{--} are the coefficients of right-circular to right-circular reflection and left-circular to left-circular reflection, respectively. In terms of material parameters, one has

$$\theta_K = \Im\left[\frac{\tilde{n}_+ - \tilde{n}_-}{\tilde{n}_+ + \tilde{n}_-}\right] \approx -\frac{4\pi}{\omega}\Im\left[\frac{\sigma_{xy}}{\bar{n}\left(\bar{n}^2 - 1\right)}\right] \tag{2}$$

where $\bar{n} = \frac{1}{2}(\tilde{n}_+ + \tilde{n}_-)$ and $\boldsymbol{\sigma}$ the AC complex conductivity tensor at frequency ω. In this latter form it is easier to see explicitly that θ_K is a measure of TRSB, as generally σ_{xy} is nonzero only if TRS is broken [13]. It is worth emphasizing that PKE is strictly a consequence of broken TRS in any material with dissipation – that is, in any real system – regardless of the underlying microscopic origin of the effect [18,19]. Moreover, PKE is, with relatively few exceptions [20], generated by TRSB order of a ferromagnetic type in the sense of having an associated static, net moment.

As can be seen from Eq. (2), the question of how PKE arises is essentially answered by identifying mechanisms for generating anomalous Hall effect (AHE). Currently, two classes of models can account for AHE/PKE originating from the order parameter symmetry of a TRSB superconductor. In the first, PKE may result from skew-scattering of chiral Cooper pairs off of impurities, as a consequence of broken translational symmetry [21,22]. In the second, PKE may be generated in the clean limit if chiral pairing takes place across multiple bands [23,24]. In either case, TRSB – and therefore PKE – is only possible if the order parameter has multiple independent components that carry a relative phase between them (*e.g.* $k_x + ik_y$, $k_z(k_x + ik_y)^2$, *etc.*). Such an arrangement is only allowed if the order parameter belongs to a two- (or higher) dimensional representation of the point group [25,26].

3. Measurement of weak PKE signals

The Kerr rotation from a TRSB superconductor is estimated to be $\theta_K \approx \frac{v_F}{c}\frac{\xi}{\lambda}\frac{\Delta}{E_F}\ln(\frac{E_F}{\Delta}) \lesssim 1$ µrad, where v_F, E_F are the Fermi velocity and energy, ξ is the coherence length, and λ is the wavelength of the incident light, taken to be off-resonance with Δ [17]. Such signals are well below the resolution of standard crossed-polarizer measurement techniques (see for example [27]). Furthermore, the signal-to-noise ratio is limited by the requirement that the incident power on the sample not exceed, at best, some tens of µW, owing to the significance of optical sample heating at the sub-Kelvin temperatures at which the physics relevant to these studies occur.

The need to resolve such small signals at such low power poses steep instrumentation challenges. As a result, the zero-area loop Sagnac interferometer was developed, in which competing geometric and other interference effects are largely eliminated by routing the counterpropagating branches of the interferometer through the two orthogonal modes of a single strand of polarization-maintaining fiber [15,28,29]. In this review, experiments used wide-bandwidth light centered at $\lambda = 1550$ nm ($\sim$ 0.8 eV) and focused to a diameter of $d \sim 10.6$ µm on the sample. With this configuration, the sensitivity of the instrument is $\sim$ 10–20 nrad, depending somewhat on the reflectivity of the sample.

The PKE measurements that follow share a common framework to determine whether a given superconductor breaks TRS. A sample is cooled through T_c, usually to the base temperature (300 mK) of the cryostat, either with (field-cooled, FC) or without (zero-field-cooled, ZFC) a magnetic field applied normal to the sample surface; for FC measurements the field is switched off at the lowest temperature. All of the data shown are then recorded in zero field during a controlled warmup of the sample through T_c. Under these conditions, a TRSB superconducting order parameter is reflected by the following features:

1. **A nonzero Kerr rotation** $\Delta\theta_K$ onsets at T_c**.***
2. **The sign and magnitude of** $\Delta\theta_K$ **may vary after each cooldown under ZFC conditions**, due to the formation of randomly oriented TRSB domains. *In cases where the domains are substantially smaller than the probe beam spot size, the net PKE may cancel out under zero-field cooling conditions.
3. Conversely, the application of a small training field along the optical axis as the sample is cooled through T_c should suffice to align all of the domains. When the field is turned off well below T_c, the domains should remain aligned. Thus **$\Delta\theta_K$ is fully saturated upon (zero-field) warmup after cooling through T_c in a small training field, with the sign of $\Delta\theta_K$ determined by the sign of the training field.** In addition, the largest ZFC signal should be no larger than the FC signal: $|\Delta\theta_K^{ZFC}| \leq |\Delta\theta_K^{FC}|$.
4. As a corollary, **the magnitude of** $\Delta\theta_K$ **does not depend on the magnitude of the training field, for low field strengths.** This is an important difference between PKE originating from a chiral order parameter and PKE originating from trapped magnetic flux. In the latter case the density of vortices increases when larger fields are applied; hence, in the study of chiral superconductors it is preferable to minimize any potential vortex contribution to $\Delta\theta_K$ by applying only weak training fields.
5. **The temperature evolution of** $\Delta\theta_K^{FC}$ **roughly follows the Ginzburg–Landau expression for the product of the real and imaginary components of the gap.** In contrast, the temperature evolution of a magnetic signal generated by trapped flux rises sharply as $T \to 0$ [30]. If there is only one superconducting transition, as in unstrained Sr_2RuO_4, all of the gap components have the same T_c, and $\Delta\theta_K(T) \propto |\Delta|^2 \propto 1 - (T/T_c)^2$. However, the full expression

$$\Delta\theta_K(T) \propto |\Re(\Delta)\cdot\Im(\Delta)| \propto \sqrt{1 - \left(\frac{T}{T_{c,\Re}}\right)^2}\cdot\sqrt{1 - \left(\frac{T}{T_{c,\Im}}\right)^2} \tag{3}$$

allows for cases where the transition temperatures of the two gap components $T_{c,\Re}$ and $T_{c,\Im}$ are not necessarily identical, as in UPt_3 [29,31] and strained Sr_2RuO_4 [32].

The above criteria are applied in each of the examples that follow to determine whether the order parameters in question break TRS.

4. Results

4.1. Cecoin$_5$

The single-layer tetragonal compound $CeCoIn_5$ has, at $T_c =$ 2.3 K, the highest transition temperature of the Ce-based heavy