Intrinsic Optical Dichroism in the Chiral Superconducting State of Sr_2RuO_4

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We present an analysis of the Hall conductivity $\sigma_{xy}(\omega, T)$ in time reversal symmetry breaking states of exotic superconductors. We find that the dichroic signal is nonzero in systems with interband order parameters. This new intrinsic mechanism may explain the Kerr effect observed in strontium ruthenate and possibly other superconductors. We predict coherence factor effects in the temperature dependence of the imaginary part of the ac Hall conductivity $\mathrm{Im}\sigma_{xy}(\omega, T)$, which can be tested experimentally.

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A superconducting state with chiral p-wave symmetry is of general interest because it is the charged analogue of the superfluid A phase of ^{3}He. In such a state the Cooper pairs are spin triplets and have a relative angular momentum $l = 1$, and therefore it occupies a unique place on the list of superfluid phases of matter. Furthermore, this particular state has been identified recently as a possible topological superconductor emphasizing its relevance to understanding of superfluidity at the deepest level [1]. The best candidate to host this exotic state of matter appears to be Sr_2RuO_4 [2]. The central supporting evidence for chiral p-wave symmetry in this material is provided by experiments which show that the superconducting state breaks time reversal invariance [3,4].

The possibility of using optical dichroism to detect time reversal symmetry breaking (TRSB) pairing states in unconventional superconductors was first suggested in the late 1980s [5–8]. Recently, such dichroism was observed in polar Kerr effect measurements of the 1.5 K superconductor Sr_2RuO_4 by Xia *et al.* [9]. Subsequently, similar dichroism was found in some underdoped high temperature superconductors [4]. The measurements on strontium ruthenate showed a small Kerr rotation of light of wavelength $\lambda = 1550$ nm, corresponding to a rotation of the plane of polarization by an amount approaching 100 nrad at $T = 0$ and going to zero at T_c approximately linearly in $T_c - T$. Strong evidence for TRSB in strontium ruthenate had previously been seen in muon spin rotation [3], where the signal shows a broadly similar temperature dependence. Together these observations support the identification of this material as a chiral p-wave superconductor [2]. However, the theoretical interpretation of both of these experiments is difficult and edge currents predicted by the chiral pairing theory have not been observed [10,11], leaving the question of the identification of the pairing state partially unresolved [12].

In particular, the origin of dichroism in a chiral superconducting state has attracted considerable attention in the recent literature [13–19]. The conclusion of this work is that the dichroic signal is exactly zero in the intrinsic limit, and only appears as a higher order effect in the presence of impurity scattering [17–19]. Numerical estimates of the Kerr signal arising from this mechanism appear consistent with the experimental observations [4].

In this Letter we propose a different mechanism for the generation of the dichroic signal, which is purely intrinsic and does not rely on impurity scattering or a finite width of the incident photon beam. The principal difference between this work and the earlier calculations is that our theory is based upon a multiband pairing model of Sr_2RuO_4, and, as we show below, the dichroic signal arises from interorbital pairing associated with the d_{xz} and d_{yz} Ru orbitals. We have previously shown that this same model gives a good description of the overall thermodynamic properties of Sr_2RuO_4 [20–22]. Crucially, the same interorbital pairing model predicts a finite orbital magnetic moment on each Ru atom [23], which has the same origin as the calculated dichroic signal. The two are in fact directly linked by the f-sum rule [24]. The fact that interorbital pairing associated with the Ru d_{xz} and d_{yz} is the key physical feature of dichroism in this theory is qualitatively consistent with the proposals by Raghu, Kapitulnik, and Kivelson [25]; however, in our phenomenological theory all bands are assumed to be superconducting with comparable values of the gap [20].

Our calculation of the optical dichroism is based on the systematic analysis of the Bogoliubov–de Gennes (BdG) equations developed by Capelle, Gross, and Gyorffy [26,27]. They discuss a fairly complete list of conditions, including TRSB, under which dichroism in the electromagnetic response of a superconductor occurs. In this formalism the conductivity tensor can be expressed in terms of the electromagnetic power absorption $P(\omega, \boldsymbol{\epsilon})$ for light of left and right circular polarizations, $\boldsymbol{\epsilon}_L$ and $\boldsymbol{\epsilon}_R$, respectively,

$$\mathrm{Im}[\sigma_{xy}(\omega)] = \frac{1}{VE_0^2}[P(\omega, \boldsymbol{\epsilon}_L) - P(\omega, \boldsymbol{\epsilon}_R)]. \quad (1)$$

Here V is the sample volume, E_0 is the electric field strength of the light, and $\boldsymbol{\epsilon}_{L/R} = (1, \pm i, 0)/\sqrt{2}$. Within the BdG formalism the absorption spectrum can be calculated directly in terms of the dipole matrix elements [26,27]

$$P(\omega, \boldsymbol{\epsilon}) = \frac{\pi^2 e^2 E_0^2}{2\omega} \sum_{N,N',\mathbf{k}} f(E_N(\mathbf{k}))[1 - f(E_{N'}(\mathbf{k}))] \times |\langle N'\mathbf{k}|\hat{H}_I(\boldsymbol{\epsilon})|N\mathbf{k}\rangle|^2 \delta(E_{N'}(\mathbf{k}) - E_N(\mathbf{k}) - \hbar\omega), \quad (2)$$

where

$$|\, N\mathbf{k}\rangle = \begin{pmatrix} u_N(\mathbf{k}) \\ v_N(\mathbf{k}) \end{pmatrix} \quad (3)$$

is the Nth eigenvector of the BdG equation at wave vector $\mathbf{k}$ fulfilling the equation

$$\begin{pmatrix} \hat{H}_0(\mathbf{k}) & \hat{\Delta}(\mathbf{k}) \\ \hat{\Delta}(\mathbf{k})^\dagger & -\hat{H}_0(\mathbf{k})^* \end{pmatrix} \begin{pmatrix} u_N(\mathbf{k}) \\ u_N(\mathbf{k}) \end{pmatrix} = E_N \begin{pmatrix} u_N(\mathbf{k}) \\ u_N(\mathbf{k}) \end{pmatrix}. \quad (4)$$

Here $\hat{H}_0(\mathbf{k})$ is the normal state tight-binding Hamiltonian, $\hat{\Delta}(\mathbf{k})$ is the matrix of gap parameters in the tight-binding spin-orbital basis. The matrix elements of the light-matter interaction Hamiltonian in Eq. (2) have the general form

$$\langle N'\mathbf{k} \mid \hat{H}_I \mid N\mathbf{k}\rangle = (u^*_{N'}(\mathbf{k}), v^*_{N'}(\mathbf{k})) \begin{pmatrix} \boldsymbol{\epsilon}\cdot\hat{\mathbf{v}} & 0 \\ 0 & -(\boldsymbol{\epsilon}\cdot\hat{\mathbf{v}})^* \end{pmatrix} \begin{pmatrix} u_N(\mathbf{k}) \\ v_N(\mathbf{k}) \end{pmatrix}, \quad (5)$$

where $\hat{\mathbf{v}} = \nabla_{\mathbf{k}}\hat{H}_0(\mathbf{k})/\hbar$ is the velocity operator. In the tight-binding representation of the Sr_2RuO_4 bands [23], the wave functions are $u_N(\mathbf{k}) \equiv u_N^{m\sigma}(\mathbf{k})$ and $v_N(\mathbf{k}) \equiv v_N^{m\sigma}(\mathbf{k})$, where the orbital index m runs over the three Ru $4d$ orbitals (d_{xz}, d_{yz}, d_{xy}) and the index σ represents electron spin. In this basis $\hat{H}_0(\mathbf{k})$ is the 6×6 tight-binding Hamiltonian, including both on-site energies, hopping integrals, and, in general, spin-orbit interactions. Most of the calculations described below have been performed for the set of parameters used earlier [21] in our modeling of strontium ruthenate with nonzero out-of-plane interorbital interactions between d_{xz} and d_{yz} orbitals.

We start the discussion by showing in Fig. 1 the temperature dependence of the imaginary part of the three dimensional Hall conductivity $\mathrm{Im}\sigma_{xy}(T, \omega)$ calculated for a number of frequencies ω. Note that the results have been shown in natural units for three dimensional conductivity, i.e., $\frac{e^2}{hd}$, where e is the electron charge h, Planck's constant, and d the c-axis lattice constant, $d = 1.3$ nm in strontium ruthenate. The energies are measured in units of t, the in-plane hopping parameter between d_{xy} orbitals, which has been estimated to be $t = 0.081\,62$ eV.

In Fig. 1 the frequencies ω_0 range from smaller than the zero temperature energy gap $\Delta(0) \approx 0.0033t$ in the d_{xz} and d_{yz} orbital space, to larger than it. In the low frequency case a coherence peak is observed, which is absent for higher optical frequencies. The temperature dependence of $\mathrm{Im}\sigma_{xy}$ is easily related to that of the superconducting gap in the large frequency limit were it scales approximately as second power of the gap. The curves normalized to their low temperature values are shown in Fig. 2. It is worth noting that while the high frequency signal scales roughly as the square of normalized order parameter, the low frequency results show strong deviations, which can be identified as a coherence peak similar to the Hebel-Slichter [28] peak observed in NMR experiments on classic superconductors. This coherence peak in the temperature dependence of the dichroic signal is not apparent in the experiment [4,9], which was in the high frequency limit. For this system the observation of the coherence peak would require usage of light with low frequencies of the order $\omega_{\mathrm{cp}} \approx 0.003t = 0.245$ meV, i.e., in the far infrared region of the spectrum.

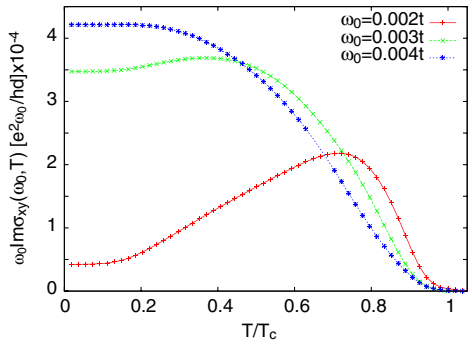

FIG. 1 (color online). The temperature dependence of the dichroic signal in the chiral state calculated for a few values of the probing light frequencies. For this particular set of interaction parameters $T_c = 0.001\,35t$, which is slightly lower than $0.0015t$ corresponding to $T_c \approx 1.5$ K.

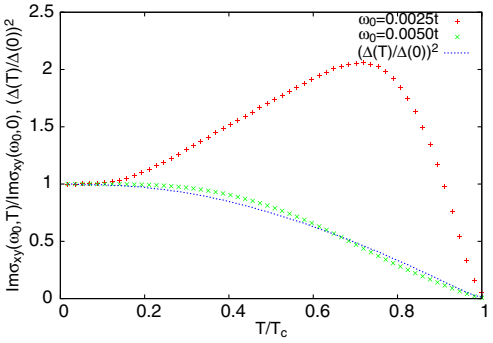

FIG. 2 (color online). The temperature dependence of the $\mathrm{Im}\sigma_{xy}(\omega_0, T)$ normalized to its low temperature value in the chiral state for two values of the light frequency: slightly below the zero temperature gap value $\omega_0 = 0.0025t$ and above it $\omega_0 = 0.0050t$ compared to the normalized gap $(\frac{\Delta(T)}{\Delta(0)})^2$. Note the roughly quadratic dependence of the dichroic signal on the gap for the probing frequency larger than the gap, and the strong departures from such a behavior for low optical frequencies.

The reflection coefficient $|r|$ and the polar Kerr angle θ_K are given by the following equations [16,29]:

$$|r| = \frac{|n-1|}{|n+1|}, \tag{6}$$

$$\theta_K = \frac{4\pi}{\omega}\,\mathrm{Im}\frac{\sigma_{xy}(\omega)}{n(n^2-1)}, \tag{7}$$

where n is the complex refraction coefficient. The polar Kerr angle (7) has been found [16] in the high frequency regime ($\omega > \omega_{ab}$) to read

$$\theta_K = \frac{4\pi\omega^2\,\mathrm{Im}\sigma_{xy}(\omega)}{\sqrt{\epsilon_\infty\omega^2-\omega_{ab}^2}[(\epsilon_\infty-1)\omega^2-\omega_{ab}^2]} \tag{8}$$

and

$$\theta_K = -\frac{4\pi\omega^2\,\mathrm{Re}\sigma_{xy}(\omega)}{\sqrt{\omega_{ab}^2-\epsilon_\infty\omega^2}[(\epsilon_\infty-1)\omega^2-\omega_{ab}^2]}, \tag{9}$$

for light frequencies smaller than in-plane plasma frequency ω_{ab}.

The frequency dependence of the $\mathrm{Im}\sigma_{xy}(\omega)$ is shown in Fig. 3 for low frequencies and temperature close to 0 K.

The approach we use here gives us an access to the elements $\mathrm{Im}\sigma_{xy}(\omega, T)$ and $\mathrm{Re}\sigma_{xx}(\omega, T)$ of the conductivity tensor. To calculate $\mathrm{Re}\sigma_{xy}(\omega)$ needed to calculate θ_K in the frequency limit appropriate for experiments ($\Delta \ll \omega < \omega_{ab}$) one has to perform Kramers-Kronig analysis [30]. To this end the full frequency dependence of the $\mathrm{Im}\sigma_{xy}(\omega)$ is needed. Assuming that at very high frequencies $\omega\mathrm{Im}\sigma_{xy}(\omega)$ tends to a constant we obtain $\mathrm{Re}\sigma_{xy}(\omega = 0.8\ \mathrm{eV} = 9.8t) \approx 1.8\times10^{-6}$ in natural units $e^2/(hd)$. This number together with the approximation

$$\theta_K = 4\pi\frac{\omega^2}{\omega_{ab}^3}\mathrm{Re}\sigma_{xy}(\omega), \tag{10}$$

and the experimental value of plasma frequency $\omega_{ab} = 4.5\ \mathrm{eV} \approx 55.1t$ gives $\theta_K \approx 200$ nrad, which is reasonably close to the experimental value of order of 90 nrad.

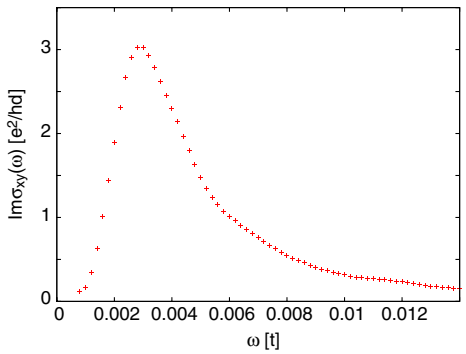

FIG. 3 (color online). The frequency dependence of $\mathrm{Im}\sigma_{xy}$ calculated for the chiral state at low temperature.

The dichroic signal we obtain from the imaginary part of the Hall conductivity changes sign with chirality of the state $\sin k_x \pm i\sin k_y$ and, as expected, equals exactly zero for nonchiral states. In the normal state the appearance of the dichroic signal requires both spin-orbit coupling and an external magnetic field breaking time reversal symmetry [27].

In the present calculations the nonzero dichroic signal we obtain for the chiral state of Sr_2RuO_4 can be shown to arise from interorbital (d_{xz}, d_{yz}) Cooper pairs. The signal becomes zero if we remove the pairing interaction for these interorbital pairs in our model, leaving only d_{xy} orbital pairing on a single sheet of Fermi surface. Using single band models Lutchyn *et al.* [16,18] and Goryo [17,19] have found a nonzero Kerr effect only by considering the scattering of carriers by impurities, and therefore this is an extrinsic Kerr effect. In a very clean system, like strontium ruthenate, this third order impurity scattering might seem improbable to be solely responsible for the measurements. In a very recent paper Taylor and Kallin [31] have also proposed a very similar theory for an intrinsic interband contribution to the Kerr effect in Sr_2RuO_4.

An experimental test of our mechanism is possible because the temperature dependence of the Hall conductivity is not universal. In our mechanism it shows a coherence peak similar to that found by Hebel and Slichter in the temperature dependence of nuclear relaxation time $1/T_1$ as measured in NMR. This prediction [27] can, in principle, be tested experimentally by changing the frequency of the light. This would allow the present mechanism to be compared to other possible sources of dichroism, either arising from collective excitations [5,6,8] or high order impurity scattering [16–19]. Interestingly, the presence of multiple bands around the Fermi energy occurs for many superconductors and in these systems the presence of at least a small interorbital-interband contribution to the pairing is very likely. Thus the mechanism which we propose may be operative not only in Sr_2RuO_4 but also in other systems, such as some high temperature superconductors [4].

Finally, it is of interest to recall that for normal systems the integral

$$\langle \mathrm{Im}[\sigma_{xy}(\omega)]\rangle \equiv \int_0^\infty \mathrm{Im}[\sigma_{xy}(\omega)]d\omega. \tag{11}$$

is related to a certain component of the orbital magnetization $\vec{M}$ by the f-sum rule. This was first derived by Oppeneer [32] and further discussed by Souza and Vanderbilt [24]. Clearly, if a similar relation held for superconductors it could lead to new insights into the highly controversial question of what is the total orbital momentum of a p-wave superconductor. Indeed, using (11) and following the arguments of Souza and Vanderbilt we find

$$\langle \mathrm{Im}[\sigma^z_{xy}(\omega)]\rangle = \frac{\pi^2 e^2}{V}(\mathrm{tr}[\hat{P}_{u,u}\hat{r}\times\hat{Q}_{u,u}\hat{v}] - \mathrm{tr}[\hat{P}_{v,v}\hat{r}\times\hat{Q}_{v,v}\hat{v}])_z + \Sigma^z_{x,y}, \quad (12)$$

where $\hat{r}$ and $\hat{v}$ are the position and velocity operators, respectively, and the particle and hole projection operators are defined as $\hat{P}_{u,u} = \sum_N |u_{N'}\rangle f_N \langle u_N|$ and $\hat{Q}_{u,u} = \sum_N |u_{N'}\rangle(1-f_N)\langle u_N|$, respectively, with f_N the Fermi Dirac distribution of the quasiparticle state of energy $E_N(\mathbf{k})$. In Eq. (12) the contribution $\Sigma^z_{x,y}$ is given by

$$\Sigma_{x,y} = \frac{\pi^2 e^2}{2V}\sum_{N,N'}\{f_N x^{u,u}_{N,N'}(1-f_{N'})v^{v,v}_{y;N'N} - f_N y^{v,v}_{N,N'}(1-f_{N'})v^{u,u}_{x;N'N} - f_N x^{v,v}_{N,N'}(1-f_{N'})v^{u,u}_{y;N'N} + f_N y^{u,u}_{N,N'}(1-f_{N'})v^{v,v}_{x;N'N}\}, \quad (13)$$

where, for brevity, we use the notation $x^{u,u}_{N,N'} \equiv \langle u_N \mid x \mid u_{N'}\rangle$, $v^{v,v}_{y;N'N} \equiv \langle v_{N'} \mid v_y \mid v_N\rangle$, etc.

The first term in Eq. (12) is a contribution to the total angular momentum given by the particles and holes separately. One may regard it as a quasiparticle contribution to the orbital magnetization. Reassuringly, in the normal state it reduces to the component of the orbital magnetization defined by Souza and Vanderbilt [24] as M_{RS}. On the other hand, the second contribution in Eq. (12), $\Sigma^z_{x,y}$, as can be seen in Eq. (13), involves products of both particle and hole amplitudes and therefore can be regarded as the consequence of the order parameter, namely, the condensate. Further discussion of this very interesting f-sum rule will be published elsewhere [33].

Here we merely note that the f-sum rule for Sr_2RuO_4, shown in Fig. 4, also has a characteristic temperature dependence, which can be compared with experiments and with other theories of orbital magnetization in the chiral pairing state. For example, we can compare this temperature dependence to that which we calculated previously [23] for the same tight-binding Hamiltonian and model gap equation for Sr_2RuO_4 as discussed in this Letter. We previously estimated that the orbital magnetization M^z_{RS} in the chiral superconducting state had a temperature dependence which fitted very well with that of $(\frac{\Delta(T)}{\Delta(0)})^2$. It is clear from Fig. 4 that this gives a reasonable, but not perfect, fit to the results obtained from the f-sum rule. The previous calculation [23] evaluated the magnetization in a theory which only included the first, quasiparticle, terms in Eq. (12). Thus we attribute the corresponding deviation in Fig. 4 to the contribution of the condensate terms, Eq. (13). This suggests that the mechanism of dichroism arising from interorbital pairing discussed in this Letter operates through both the quasiparticle excitations and the condensate to produce the total contributions to the dichroic signal.

In conclusion, we predict the existence of an intrinsic dichroic signal in systems with interorbital-interband Cooper pairs with chiral symmetry of the order parameter. These calculations also suggest that a nonzero Hall conductivity may also arise in other materials having intraorbital order parameters, with differing phases of the order parameters between the various orbitals. In this case the interorbital order parameter derived from the d_{xz} and d_{yz} Ru orbitals is ultimately responsible for the effect.

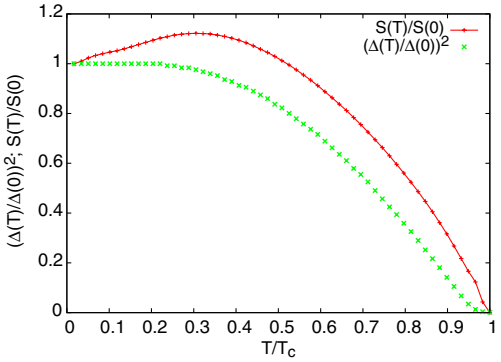

FIG. 4 (color online). The temperature dependence of the f-sum rule $S(T) \equiv \langle \mathrm{Im}[\sigma^z_{xy}(\omega)]\rangle$ normalized to its zero temperature value $S(0)$. This is compared with the square of normalized order parameter $(\Delta(T)/\Delta(0))^2$.

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