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Theory of dichroism in high-temperature superconductors

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We consider the circumstances under which non-s-wave superconductivity could give rise to optical dichroism. Depending on the type of state, circular or linear dichroism and birefringence can occur, and we show how the experiments can be used to investigate the microscopic pair wave function. We also show that we can reconcile the seemingly contradicting experimental results by assuming spin-orbit coupling and the mixing of spin singlet and triplet.

Much recent research has focused on the possibility of broken time-reversal (T) and parity (P) symmetry in high-temperature superconductors. This arose from the anyon model[1,2] of these materials, in which the ground-state breaks T and P.[3] This model is justified by the analogy to the fractional quantum Hall effect. Since recent muon spin-resonance experiments[4] seem to be inconsistent with anyon theories (in which charge and flux are always bound together), it makes sense to look at alternative ways of breaking T and P. It has been known for some years that the ground state of heavy-fermion superconductors may also break these symmetries. Thus the present work is motivated theoretically by the analogy to heavy-fermion superconductivity.

Recent work searching for circular dichroism in high-T_c materials gave the main experimental motivation for the work presented here. Positive evidence for circular dichroism has been reported in two papers.[5,6] Negative results for a nonreciprocal Faraday effect have also been reported.[7] We present an analysis of these observations based on various possible forms of unconventional superconductivity.

There are experimental grounds for supposing that the superconducting order parameter may be a mixture of s- and d-wave pairing, even before the optical experiments. In particular, a picture in which the pairing is predominantly d-wave near T_c and cross over to a mixture of low temperatures is suggested by earlier experiments. The absence of a Hebel-Slichter peak near T_c in the relaxation rate of nuclear magnetic moments[8] is characteristic of unconventional superconductivity. Other evidence has been summarized by Annett *et al.*[9] On the other hand, the weak temperature dependence of the penetration depth at temperatures much less than T_c indicates a nodeless gap function with an s-wave component, through this conclusion has been recently challenged.[10]

The dielectric tensor is given by

$$\epsilon_{\alpha\beta}(\mathbf{q},\omega)=\epsilon^0_{\alpha\beta}+\frac{4\pi i}{\omega}\sigma_{\alpha\beta}(\mathbf{q},\omega)\,,$$

where the second term is the contribution of the itinerant electrons. In the clean limit of weak-coupling BCS theory, we may write this term as

$$\sigma_{\alpha\beta}(\mathbf{q},\omega)=\frac{i}{\omega}\left[\Pi_{\alpha\beta}(\mathbf{q},\omega)+\frac{ne^2}{m}\delta_{\alpha\beta}\right],$$

where $\Pi_{\alpha\beta}(\mathbf{q},\omega)$ is the current-current correlation function,

$$\Pi_{\alpha\beta}(\mathbf{q},\omega)=\frac{1}{4\pi^3}\int d^3p[j_{\alpha\uparrow}(\mathbf{p})j_{\beta\downarrow}(\mathbf{p})F(\mathbf{p},\mathbf{q},\Delta)+j_{\alpha\uparrow}(\mathbf{p})j_{\beta\uparrow}(\mathbf{p})G(\mathbf{p},\mathbf{q},\Delta)]\,. \quad (1)$$

The symbols appearing in Eq. (1) are defined as

$$F(\mathbf{p},\mathbf{q},\Delta)=[1-n_F(E_{p_+})-n_F(E_{p_-})]F_1+[n_F(E_{p_-})-n_F(E_{p_+})]F_2\,,$$

$$G(\mathbf{p},\mathbf{q},\Delta)=[1-n_F(E_{p_+})-n_F(E_{p_-})]G_1+[n_F(E_{p_-})-n_F(E_{p_+})]G_2\,,$$

where

$$F_1=-\frac{\Delta(\mathbf{p}_+)\Delta^*(\mathbf{p}_-)}{4E_{p_+}E_{p_-}}\left[\frac{1}{\omega-E_{p_+}-E_{p_-}+i\gamma}-\frac{1}{\omega+E_{p_+}+E_{p_-}+i\gamma}\right],$$

$$F_2=\frac{\Delta(\mathbf{p}_+)\Delta^*(\mathbf{p}_-)}{4E_{p_+}E_{p_-}}\left[\frac{1}{\omega-E_{p_+}+E_{p_-}+i\gamma}-\frac{1}{\omega+E_{p_+}-E_{p_-}+i\gamma}\right],$$

$$G_1=\left[\frac{u^2_{p_+}+v^2_{p_-}}{\omega-E_{p_+}-E_{p_-}+i\gamma}-\frac{u^2_{p_-}v^2_{p_+}}{\omega+E_{p_+}+E_{p_-}+i\gamma}\right],$$

$$G_2=\left[\frac{u^2_{p_+}u^2_{p_-}}{\omega-E_{p_+}+E_{p_-}+i\gamma}-\frac{v^2_{p_+}v^2_{p_-}}{\omega+E_{p_+}-E_{p_-}+i\gamma}\right].$$

Here

$$u_p^2=\tfrac{1}{2}(1+\xi_p/E_p),\quad v_p^2=\tfrac{1}{2}(1-\xi_p/E_p)\,,$$

$$E_p^2=|\Delta(\mathbf{p})|^2+\xi_\mathbf{p}^2,\text{ and }\mathbf{p}_\pm=\mathbf{p}\pm\mathbf{q}/2\,.$$

$j_{\alpha\uparrow}(\mathbf{p})=\langle\mathbf{p}\uparrow|\hat{\mathbf{j}}_\alpha|\mathbf{p}\uparrow\rangle$ is the matrix element of the current operator between eigenstates of the crystal Hamiltonian and $n_F(E)$ is the Fermi distribution function.

We can draw some useful conclusions about $\Pi_{\alpha\beta}$ and therefore $\epsilon_{\alpha\beta}$ from Eq. (1) without evaluating the integral, and without assuming any particular form for $\Delta(\mathbf{p})$. Let us assume that the system is tetragonal and is exposed to light incident along the z direction so that $\mathbf{q}=q\hat{\mathbf{z}}$. In the normal state, which we assume to be a Fermi liquid, $\Delta=0$ and $\Pi_{xy}=\Pi_{yx}=\epsilon_{xy}=\epsilon_{yx}=0$. The function $j_{\alpha\uparrow}(\mathbf{p})j_{\beta\downarrow}(\mathbf{p})$ belongs to the Γ_4 representation of C_{4v}. (We use the same notation as in Ref. 11.) In Table I we list the possible mixings of the s- and d-wave pairing states in the tetragonal lattice. Here we assume the s-wave state in its simplest form, i.e., a constant, but the conclusions would be unchanged if it were replaced with the extended s-wave $(k_x^2+k_y^2)$. We can see that $\Delta(\mathbf{p})$ must belong to one of the mixed representations $\Gamma_1+\Gamma_4$, $\Gamma_2+\Gamma_3$, or the pure representation Γ_5 in order that $\Pi_{xy}\neq 0$. Among them only $\Gamma_1+\Gamma_4$ is possibly nodeless, while $\Gamma_2+\Gamma_3$ or Γ_5 gap functions must have nodes and are therefore presumably ruled out experimentally. In what follows, therefore, we concentrate on the $\Gamma_1+\Gamma_4$ case. Explicitly, this corresponds to a gap function of the form, e.g.,

$$\Delta(\mathbf{k})=\Delta_s(T)f_s(k_z)+\Delta_d(T)\sin k_x \sin k_y f_d(k_z)\ , \quad (2)$$

where f_s and f_d are even functions of k_z in the spin singlet case. We can always assume f_s to be real because of the U(1) gauge symmetry. $\Delta_s(T)$ and $\Delta_d(T)$ are real functions of temperature and $\Delta_d(T)$ is taken to dominate near T_c. If f_d is an imaginary function, at low temperatures we would still find $|\Delta|>|\Delta_s|$ for all $\mathbf{k}$, i.e., a nodeless energy-gap function. We shall call the gap function of Eq. (2) with an imaginary f_d the $s+id$ state hereafter.

Theoretically, a mixture of s- and d-like functions has been found to be a favorable ground state for the t-J model in mean field[12] and variational calculations.[13] The latter, in particular, have confirmed that the relative imaginary phase between the s- and d-components is preferred.

Inserting Eq. (2) into Eq. (1), we find a dielectric tensor

$$\epsilon_{\alpha\beta}(\mathbf{q},\omega)=\begin{pmatrix}\epsilon_{xx} & \epsilon_{xy} & 0\\ \epsilon_{yx} & \epsilon_{yy} & 0\\ 0 & 0 & \epsilon_{zz}\end{pmatrix} .$$

If the system does not break time-reversal symmetry T but only breaks parity P [$f_d(k_z)$ is a real function], we have $\epsilon_{xy}=\epsilon_{yx}$ from Eq. (1). ϵ_{xy} is, in general, a complex number. This equality of off-diagonal components leads to *linear* dichroism and linear birefringence with the optical axes along $\hat{\mathbf{n}}_1=(1/\sqrt{2},1/\sqrt{2},0)$ and $\hat{\mathbf{n}}_2=(1/\sqrt{2},-1/\sqrt{2},0)$. (The z axis is of course always an optical axis.) This will cause a rotation of the polarization plane if the incident linearly polarized light is not along one of the optical axes in a transmission or reflection experiment. However, this kind of signal was rejected in the experiment of Ref. 5. This was accomplished by rotating the incident polarization plane and then averaging the output.

TABLE I. Possible mixings of s- and d-wave pairing states in the tetragonal lattice. In the last column "no" should be understood as possibly nodeless.

	Form	Π_{xy}	$\Pi_{xx}-\Pi_{yy}$	Node
$\Gamma_1+\Gamma_2$	$\Delta_s+\Delta_d k_x k_y(k_x^2-k_y^2)$	0	0	no
$\Gamma_1+\Gamma_3$	$\Delta_s+\Delta_d(k_x^2-k_y^2)$	0	$\neq 0$	no
$\Gamma_1+\Gamma_4$	$\Delta_s+\Delta_d k_x k_y$	$\neq 0$	0	no
Γ_5	$\Delta_1 k_x+\Delta_2 k_y$	$\neq 0$	$\neq 0$	yes
$\Gamma_2+\Gamma_3$	$(\Delta_1 k_x k_y+\Delta_2)(k_x^2-k_y^2)$	$\neq 0$	0	yes
$\Gamma_2+\Gamma_4$	$[\Delta_1(k_x^2-k_y^2)+\Delta_d]k_x k_y$	0	$\neq 0$	yes
$\Gamma_3+\Gamma_4$	$\Delta_1(k_x^2-k_y^2)+\Delta_2 k_x k_y$	0	0	yes

If the system does break T such as in the case of $s+id$ state of Eq. (2) with an imaginary f_d, then we have

$$\Pi_{xy}(\Delta)=\Pi_{yx}(\Delta^*) \quad (3)$$

according to Eq. (1). This is actually an example of an *Onsager relation* for the T broken case.[14] Δ^* is the time-reversed state of Δ. When we evaluate Π_{xy} of $s+id$ state using Eq. (1), we find that only the integral of the F term is nonzero. The F term is proportional to

$$\Delta(\mathbf{p}_+)\Delta^*(\mathbf{p}_-)=\Delta_s^2 f_s(p_z+q)f_s(p_z)+\Delta_d^2\sin^2 p_x\sin^2 p_y f_d(p_z+q)f_d^*(p_z)$$
$$+\Delta_s\Delta_d\sin p_x\sin p_y[f_s(p_z)f_d(p_z+q)+f_s(p_z+q)f_d^*(p_z)]\ , \quad (4)$$

in which only the last term survives the p_x,p_y integrals. Furthermore, we also find that the p_z integral of the last term of Eq. (4) is zero because $f_d(p_z)$ is a periodic and even function of p_z. We conclude that $\Pi_{xy}=0$ in any spin-singlet $s+id$ states. There is, however, another intriguing possibility, which is that the Cooper pair wave function has *both* spin singlet and spin triplet character. In this case, the gap function is a 2×2 matrix, but Eqs. (1)–(4) still give essentially the correct results. $f_d(k_z)$ is now an odd function. In the presence of spin-orbit coupling, $\langle\mathbf{p}\uparrow|\hat{\mathbf{j}}_\alpha|\mathbf{p}\downarrow\rangle$ no longer vanishes. (The spin index must be thought as pseudospin, $|\mathbf{p}\downarrow\rangle$ being obtained by adiabatic continuation in the strength of the spin-orbit coupling from the pure spin down state.) While spin-orbit coupling is small in high-T_c materials, it is also small in the transition-metal magnets which are the usual subjects of optical rotation experiments. Now Π_{xy} is nonzero, and we have introduced helicity into the superconducting state. This gives rise to a *reciprocal* rotation effect since $\epsilon_{xy}=iaq$ and $\epsilon_{yx}=-iaq$ would be proportional to $\mathbf{q}$. Here $\mathbf{q}$ is the wave vector of the incident light. In this case, the system will exhibit *circular* dichroism and birefringence. More accurately, in the materials studied in the experiments, the crystal symmetry is orthorhombic instead of tetragonal so that the eigenvectors of the dielectric constant tensor correspond to elliptically polarized light.

The hypothesis of unconventional superconductivity is thus qualitatively consistent with the results of the polar Kerr effect experiments[5,6] and the null result of Ref. 7. The correlation of the superconductivity and the circular dichroism observed in the experiments provides further support for the hypothesis.[15] The use of Eq. (1), based on mean-field theory, to compute the temperature dependence of the rotation angle leads to a rather poor quantitative comparison of theory and experiment, because this effect must vanish above T_c in mean-field theory, as we will see below. Inclusion of fluctuation effects by standard methods would produce a nonzero effect above T_c, however, and would go in the direction of reconciling theory and experiment.

Next we evaluate $\epsilon_{\alpha\beta}$ of the $s+id$ state of Eq. (2) using Eq. (1). We assume that $f_s(k_z)=\cos k_z$ and $f_d(k_z)=i\sin k_z$. In the Kerr effect experiment, the Kerr rotation ϕ_K and the Kerr ellipticity ϵ_K is given by[16]

$$\phi_K+i\epsilon_K=\frac{\epsilon_{xy}}{\epsilon_{xx}^{1/2}(1-\epsilon_{xx})}\,. \tag{5}$$

In the clean limit, the momentum is conserved. ϵ_{xy} is calculated to be about $0.7\times10^{-6}i$ at zero temperature. Equation (5) tells us that the Kerr ellipticity is proportional to ϵ_{xy} and $\epsilon_{xy}=10^{-6}i$ would roughly produce a rotation of 1 μrad. This is too small to explain the observed signals in the experiments of Refs. 5 and 6. In the dirty limit, however, the conservation of momentum is lifted and ϵ_{xy} is increased to about $1.1\times10^{-4}i$ which is in the same magnitude as the observed experimental results.

In Fig. 1, we show the calculated result of the rotation angle of the polarization plane as a function of the temperature in a polar Kerr effect experiment. We assume a cylindrical Fermi surface with $E_F=0.2$ eV, wavelength of the light $\lambda=6000$ Å, and critical temperature $T_c=100$ K. We also assume that both $\Delta_s(T)$ and $\Delta_d(T)$ have BCS-like temperature dependence. We use the lattice constant and the complex dielectric constant of $YBa_2Cu_3O_7$ in our calculation.[17,18] In the experiment of Ref. 7, since the polarization rotation angles depend on which direction it travels, the rotation is reciprocal which is in agreement with the experimental null result.

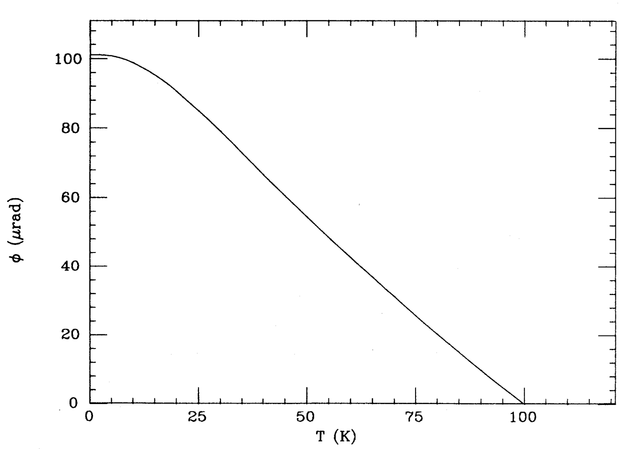

FIG. 1. Calculated Kerr rotation angle as a function of temperature using Eqs. (1) and (5). We use the parameters of $YBa_2Cu_3O_7$ in our calculation. $\epsilon_{xx}=1.85+2.35i$ at frequency $\omega=2.07$ eV is adapted from Ref. 18.

In the experiment of Ref. 6 a correlation of rotation direction to field direction on cooling was observed. So let us consider the coupling of the order parameter to an external field **H** in the z direction. The operators $p_x=-i\partial/\partial_x+2eA_x/\hbar c$ and $p_y=-i\partial/\partial_y+2eA_y/\hbar c$ belong to the Γ_5 representation. (Here **A** is the vector potential.) Because of the decomposition

$$\Gamma_1\times\Gamma_4\times\Gamma_5\times\Gamma_5=\Gamma_1+\Gamma_2+\Gamma_3+\Gamma_4\,,$$

there is a gradient term which mixes the s and d components of Δ. One finds the free energy[19]

$$\begin{aligned}\mathcal{F}=\alpha_s|\Delta_s|^2+\alpha_d|\Delta_d|^2+k_s(|p_x\Delta_s|^2+|p_y\Delta_s|^2)\\+k_d(|p_x\Delta_d|^2+|p_y\Delta_d|^2)\\+k_{sd}(p_x\Delta_s p_y^*\Delta_d^*+p_y\Delta_s p_x^*\Delta_d^*+\text{c.c.})\,.\end{aligned} \tag{6}$$

If the magnetic-field direction is reversed, $\mathbf{A}\to-\mathbf{A}$, $p_x\to-p_x^*$, and $p_y\to-p_y^*$. Clearly we expect $\Delta\to\Delta^*$. This implies $\epsilon_{xy}\to\epsilon_{xy}^*$ from Eq. (1), which leads immediately to a change in sign of the rotation angle of the polarization in the experiments. This is consistent with the experiment.[6]

The above arguments apply most simply to single-domain samples. However, it is clear from Eq. (6) that $\Delta(\mathbf{k})$ and $\Delta^*(\mathbf{k})$ are degenerate solutions to the problem in the absence of a field. Thus we expect, in general, to find some cancellation of the signal due to multidomain effects. This is observed in the experiment of Ref. 5, but seems to be absent in that of Ref. 6.

The non-s-wave superconducting order parameter's contribution to the dielectric function is not the main part in the real high-T_c materials. By assuming that those other contributions do not break the time-reversal symmetry, we single out the unconventional gap function as the only source which can produce the circular dichroism. The lack of accurate information of the value of dielectric constant matrix (due to other sources such as plasmons, phonons, etc.), especially the anisotropy in the x,y directions $\epsilon_{xx}-\epsilon_{yy}$, prevents us from making truly quantitative comparison with the experiment. However, our calculated result does agree qualitatively and semi-quantitatively with the experiments.[5-7]

Before concluding, we make some comments on the apparent discrepancy between the three experiments, namely the null result of Ref. 7 as compared to the positive results of Refs. 5 and 6. The most obvious differences are that the experiments are conducted on different samples using different frequency. Although dielectric constants of high-T_c materials do depend on frequency quite strongly, it is hard to imagine that this could produce such a big difference. The experimental setups are also quite different. Two experiments use the reflection mode, while the third one uses the transmission mode (Sagnac loop). One way to reconcile the three experiments is to assume an antiferromagnetic-type of ordering of T breaking order parameter (e.g., magnetic moment) along the z direction,[20] and take into account the off-diagonal elements of the full magnetoelectric tensor, as suggested by Dzyaloshinskii.[21] A second possibility is to introduce helicity into the superconducting state as we proposed in this paper. This can be achieved by allowing $f_d(k_z)$ in Eq. (2) to be an odd function of k_z, at the same time mixing singlet and triplet components into the Cooper pair wave function. In the presence of spin-orbit coupling, transitions between the singlet and triplet components are allowed, which would give rise to *reciprocal* rotation effects.

We conclude that the hypothesis of unconventional superconductivity is a viable explanation of recent experimental investigations of the dichroic properties of high-T_c materials. We find that one of the $s+id$ states $\Gamma_1+\Gamma_4$ of mixed singlet-triplet character exhibits circular dichroism and birefringence while the $s+d$ singlet state shows linear dichroism and birefringence, and the $s+id$ singlet state shows no dichroism or birefringence effect. We calculate the Kerr ellipticity using reasonable parameters. Our calculated results agree with the polar Kerr effect experiments qualitatively.

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