Theory of Dichroism in the Electromagnetic Response of Superconductors

K. Capelle and E. K. U. Gross
Institut für Theoretische Physik, Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany

B. L. Györffy
H.H. Wills Physics Laboratory, University of Bristol, Bristol BS8 1TL, United Kingdom
(Received 26 September 1996)

A microscopic theory of dichroism in the magneto-optical properties of superconductors is presented. Four distinct mechanisms for dichroism in superconductors are identified. Two are modifications of mechanisms known from the normal state, and two are novel effects found in superconductors only. The theory is illustrated by numerical calculations for a simple model system. The interplay between relativistic symmetry breaking and superconducting coherence is found to give rise to a variety of new effects, not known from dichroism in the normal state. [S0031-9007(97)03073-1]

PACS numbers: 74.25.Gz, 74.25.Nf, 78.20.–e

A system is said to exhibit dichroism if the absorption of light depends on its polarization [1]. Usually dichroism occurs if time reversal symmetry is broken through external or internal ("exchange") magnetic fields. The former case corresponds to the Faraday or Kerr effect, while the latter case is referred to as spontaneous dichroism. In normal metals and ferromagnets these phenomena have been the subject of many theoretical and experimental investigations [1]. For superconductors, on the other hand, very little is known about mechanisms and consequences of dichroism. The objective of this Letter is to develop a systematic theory of dichroism in superconductors.

We use the Bogoliubov–de Gennes (BdG) approach to inhomogeneous superconductors in magnetic fields [2], including relativistic effects [3], and employ perturbation theory to incorporate a variety of potential sources for dichroism. As a measure of dichroism we use the difference, $\Delta P = P_{\mathrm{LHP}} - P_{\mathrm{RHP}}$, in the power absorption of light with left-handed polarization (LHP) and right-handed polarization (RHP). Using first-order time-dependent perturbation theory, the absorbed power is

$$P = \frac{2\pi}{\hbar} \sum_{f,i} |M_{fi}^{\dagger}|^2 \Delta E_{fi} f(E_i) f(-E_f) \delta(\Delta E_{fi} - \hbar\omega)\,, \tag{1}$$

where $\Delta E_{fi} = E_f - E_i$ is the energy difference between final and initial states, and $f(E)$ is the Fermi function. Note that ΔP is proportional to $\mathrm{Im}[\sigma_{xy}(\omega)]$, the imaginary part of the off-diagonal elements of the conductivity tensor [1] which govern all magneto-optical phenomena. The matrix element M_{fi} in (1) is

$$M_{fi} = \frac{-q}{mc} \langle f| \begin{pmatrix} [\frac{ic}{2}\frac{E_0}{\omega}\boldsymbol{\epsilon}] \cdot \hat{\mathbf{p}} I & 0 \\ 0 & -([\frac{ic}{2}\frac{E_0}{\omega}\boldsymbol{\epsilon}] \cdot \hat{\mathbf{p}})^T I \end{pmatrix} |i\rangle\,, \tag{2}$$

where I is the 2×2 unit matrix. The term in square brackets is the vector potential of the external light source, expressed in terms of its electric field E_0 and the polarization vector $\boldsymbol{\epsilon}$. The latter is given by $\frac{1}{\sqrt{2}}(1, \pm i, 0)^T$ for LHP and RHP, respectively. The unperturbed system is described by the spin-dependent (four-component) BdG equation (SBdGE) [2],

$$\begin{pmatrix} \hat{h} I & i\hat{\sigma}_y \Delta \\ -i\hat{\sigma}_y \Delta^* & -\hat{h}^* I \end{pmatrix} \begin{pmatrix} u_{n\sigma}(\mathbf{r}) \\ v_{n\sigma}(\mathbf{r}) \end{pmatrix} = E_{n\sigma} \begin{pmatrix} u_{n\sigma}(\mathbf{r}) \\ v_{n\sigma}(\mathbf{r}) \end{pmatrix}. \tag{3}$$

Here $E_{n\sigma} = E_n + \sigma\mu_B B$, $\hat{h}$ is the single-particle Hamiltonian $-\frac{\hbar^2\nabla^2}{2m} + v(\mathbf{r}) - \mu + \hat{\sigma}_z \mu_B B$, and $\Delta(\mathbf{r})$ is the pair potential. $u_{n\sigma}$ and $v_{n\sigma}$ are two-component spinors with entries $u_{n\sigma\tau}(\mathbf{r})$ and $v_{n\sigma\tau}(\mathbf{r})$. For spatially constant pair potential $\bar{\Delta}$, the SBdGE eigenfunctions are [2]

$$u_{n\sigma\tau}(\mathbf{r}) = \sqrt{\frac{1}{2}\left(1 + \frac{\epsilon_n}{E_n}\right)}\, \delta_{\sigma\tau} \phi_n(\mathbf{r}) =: u_n \delta_{\sigma\tau} \phi_n(\mathbf{r})\,, \tag{4}$$

$$v_{n\sigma\tau}(\mathbf{r}) = \sigma\sqrt{\frac{1}{2}\left(1 - \frac{\epsilon_n}{E_n}\right)}\, \delta_{\tau\bar{\sigma}} \phi_n(\mathbf{r}) =: \sigma v_n \delta_{\tau\bar{\sigma}} \phi_n(\mathbf{r})\,. \tag{5}$$

Here $\bar{\sigma} = -\sigma$ and ϕ_n is a normal-state eigenfunction. Of course, if we use the solutions (4) and (5) for the initial and final states in (1), ΔP vanishes identically. Indeed, as in the normal state, one obtains dichroism if in (3) one includes terms which break chiral symmetry, such as spin-orbit coupling in the presence of magnetic fields.

Recently the SBdGE were generalized to include relativistic effects on the single-particle level [3]. From this generalization, the full form of the spin-orbit operator in superconductors is known to have off-diagonal elements involving the pair potential, in addition to the well-known diagonal spin-orbit terms which contain the lattice potential. In the presence of an external magnetic field there will also be a coupling of this field to the orbital currents, in addition to the Zeeman coupling which is already included in (3). In short-coherence-length superconductors, the inhomogeneity of the pair potential will also be important. We take this into account by writing

$\Delta(\mathbf{r}) = \bar{\Delta} + \tilde{\Delta}(\mathbf{r})$, where $\bar{\Delta}$ is the average of $\Delta(\mathbf{r})$ over one unit cell and $\tilde{\Delta}(\mathbf{r})$ the deviation from this average.

We include these effects using stationary perturbation theory. The unperturbed system is taken to be a superconductor with spatially constant pair potential, described by the SBdGE (3). We now employ the first-order stationary perturbation theory, with the perturbation

$$H_1 = \begin{pmatrix} -\frac{q}{mc}\mathbf{A}\cdot\hat{\mathbf{p}}I & \tilde{\Delta}(i\hat{\sigma}_y) \\ \tilde{\Delta}^*(i\hat{\sigma}_y)^\dagger & \frac{q}{mc}(\mathbf{A}\cdot\hat{\mathbf{p}})^* I \end{pmatrix} + \frac{\hbar}{4m^2c^2} \times \begin{pmatrix} (\boldsymbol{\sigma}\cdot\nabla v \times \hat{\mathbf{p}})I & (\boldsymbol{\sigma}\cdot\nabla\Delta\times\hat{\mathbf{p}})(i\hat{\sigma}_y) \\ [(\boldsymbol{\sigma}\cdot\nabla\Delta\times\hat{\mathbf{p}})(i\hat{\sigma}_y)]^\dagger & -(\boldsymbol{\sigma}\cdot\nabla v\times\hat{\mathbf{p}})^* I \end{pmatrix}, \tag{6}$$

where $\mathbf{A}$, the vector potential of the static magnetic field, should not be confused with that of the light wave. Note that the first term, of zeroth and first order in $1/c$, describes the effects of normal currents and order parameter inhomogeneities, while the second, of order $1/c^2$, is due to spin-orbit coupling. The term which appears as an off-diagonal element of the latter will be referred to as the "anomalous spin-orbit coupling" (ASOC) to distinguish it from the conventional spin-orbit coupling (SOC), which appears as a diagonal element. Clearly, ASOC is due to the combined effects of superconductivity and spin-orbit coupling [3]. The perturbed states constructed from (3) and (6) are then used, in a second step, as *unperturbed* states with respect to the time-dependent perturbation (2). The same procedure was used successfully in studies of dichroism in the normal state [1], although, in that case, only the conventional SOC and the diamagnetic effects contribute. In what follows we study absorption due to pair breaking only, and neglect scattering from unpaired electrons [4]. For the difference ΔP in the power absorption, this procedure yields the expression [5],

$$\begin{aligned}\Delta P(\omega, T, \mathbf{A}) = {} & \frac{\pi e^2 E_0^2}{m^2\omega}\theta(\hbar\omega - 2\Delta)\sum_{nn'\sigma} p(n,n')f(-E_{n\sigma}) \\ & \times f(-E_{n'\bar{\sigma}})\delta(E_{n\sigma} + E_{n'\bar{\sigma}} - \hbar\omega) \\ & \times \mathrm{Re}\sum_m \left[\frac{p(m,n')C_{nn'}^m}{E_n - E_m}T_{m\sigma}^+ \right. \\ & \left. + \frac{l(m,n')C_{nn'}^{\bar{m}}}{E_n + E_m}T_{m\sigma}^-\right],\end{aligned} \tag{7}$$

where Re denotes the real part. Furthermore,

$$\begin{aligned} T_{m\sigma}^+ &= u_n u_m h_{\sigma\sigma}^{nm} + u_n v_m d_{\sigma\bar{\sigma}}^{nm} + v_n u_m d_{\sigma\bar{\sigma}}^{*mn} \\ &\quad - v_n v_m h_{\bar{\sigma}\bar{\sigma}}^{*nm}, \\ T_{m\sigma}^- &= u_n v_m h_{\sigma\sigma}^{n\bar{m}} - u_n u_m d_{\sigma\bar{\sigma}}^{n\bar{m}} + v_n v_m d_{\sigma\bar{\sigma}}^{*\bar{m}n} \\ &\quad - v_n u_m h_{\bar{\sigma}\bar{\sigma}}^{*nm}, \end{aligned}$$

and $p(n,n') = u_n v_{n'} - v_n u_{n'}$ and $l(n,n') = u_n u_{n'} + v_n v_{n'}$ are coherence factors [4], with the BCS amplitudes, u_n and v_n, from (4) and (5). We also defined

$$d_{\sigma\bar{\sigma}}^{nm} = \frac{\sigma\hbar^2}{4im^2c^2}\langle n|[\nabla\Delta\times\nabla]_z|m\rangle + \langle n|\tilde{\Delta}|m\rangle, \tag{8}$$

$$d_{\sigma\bar{\sigma}}^{*nm} = -\frac{\sigma\hbar^2}{4im^2c^2}\langle n|[\nabla\Delta^*\times\nabla]_z|m\rangle + \langle n|\tilde{\Delta}^*|m\rangle, \tag{9}$$

$$h_{\sigma\sigma}^{nm} = \frac{\sigma\hbar^2}{4im^2c^2}\langle n|[\nabla v\times\nabla]_z|m\rangle + \frac{iq\hbar}{mc}\langle n|\mathbf{A}\cdot\nabla|m\rangle, \tag{10}$$

$$h_{\sigma\sigma}^{*nm} = -h_{\sigma\sigma}^{nm}, \tag{11}$$

and

$$C_{nn'}^m = 2i[p_y^{\bar{n}n'}p_x^{m\bar{n}'} - p_x^{\bar{n}n'}p_y^{m\bar{n}'}] \tag{12}$$

with normal-state matrix elements of Cartesian components of the momentum operator. We have used the particle-hole convention for the signs of the energy, hence all sums are restricted to positive energies. An index $\bar{m}$ stands for the time reversed wave function $\phi_{\bar{m}} := \phi_m^*$. In the nonsuperconducting limit this formula correctly reduces to the corresponding normal-state result. We now identify and interpret four distinct mechanisms which can give rise to a nonvanishing ΔP.

Mechanism 1.—The conventional SOC, which enters (7) through $\nabla v \times \nabla$, can, just as in normal metals, produce dichroism, provided that a magnetic field is present. It follows from (7)–(12) that, in the absence of magnetic fields, spin-orbit coupling alone will not suffice.

Mechanism 2.—Anomalous spin-orbit coupling, produced by the term $\nabla\Delta\times\nabla$, too, can give rise to dichroism if magnetic fields are present. Interestingly, this term has a different temperature behavior compared to conventional SOC.

Mechanism 3.—Orbital currents, produced by $\mathbf{A}$, such as screening currents, can lead to a finite ΔP, without SOC. In normal metals this is usually smaller than the effects due to SOC.

Mechanism 4.—Even in the absence of magnetic fields and SOC, the term containing the pair potential $\tilde{\Delta}(\mathbf{r})$ is a possible source of dichroism if $\tilde{\Delta}$ is complex. This makes dichroism a potential tool for investigating unconventional order parameters.

What all four mechanisms have in common is that time reversal symmetry is broken, either by magnetic fields or by the pair potential. Mechanisms 1 and 3, which were investigated thoroughly for normal metals, are seen to be present in superconductors as well. However, they are strongly modified by the superconducting coherence factors. Mechanisms 2 and 4 are special to superconductors and not present at all in the normal state. Those mechanisms which require magnetic fields in the superconductor, namely, 1, 2, and 3, cannot take place in the bulk of the material as long as it is in the Meissner phase. They can be active in three different situations: (a) at surfaces, within the penetration depth, (b) in the vortex phase, and (c) in superconductors displaying the coexistence of magnetism and superconductivity. Mechanisms 1 and 2 are of relativistic origin. In particular, mechanism 2 constitutes the first potentially observable consequence of the ASOC term, predicted in [3]. Mechanisms 3 and 4 were previously noted and analyzed [6]. The spin-orbit related mechanisms, although known to be dominant in normal conductors, have not been investigated in superconductors.

In the following, we calculate ΔP from (7)–(12) for a simple model. This model describes a superconductor with spatially constant pair potential and sufficiently heavy elements in the lattice so that mechanism 1 becomes dominant. In this case, we can neglect the other three mechanisms. We then proceed in a manner similar to the way in which nuclear magnetic resonance [7] and optical absorption [8] are treated within BCS theory [4]. The sums over indices labeled energy eigenstates are converted into energy integrals, and the matrix elements are approximated by averages over surfaces of constant energy and then taken out of the integrals. As a consequence of (4) and (5), all these matrix elements are *normal-state* matrix elements. The signature of superconductivity is, as usual, the presence of the coherence factors, which remain under the integrals. Forming the ratio of ΔP in the superconductor to ΔP in the normal conductor, $\Delta P^S/\Delta P^N$, the matrix elements cancel. We then assume a density of states (DOS), which models the BCS-DOS but smooths out the singularity at $E = \Delta$, using a procedure similar to that of Hebel and Slichter [7]. The integrals can then be performed numerically. To be specific, we consider a superconductor with a $T = 0$ pair potential $\Delta(T = 0) = 1$ meV, a critical temperature of $T_c = 6.6$ K, near that of lead, and a BCS-type temperature dependence of the energy gap. These values and relations can easily be modified, without changing our qualitative conclusions.

In Fig. 1 we plot $\Delta P^S/\Delta P^N$ versus the applied magnetic field. At temperatures not too close to zero, the superconductor displays stronger dichroism than the normal conductor. However, repeating the calculation at $T = 0$, we find no dichroism for the fields in Fig. 1. This is physically reasonable because at $T = 0$ a magnetic field will not produce spin polarization in a superconductor, since all electrons are paired. On the other hand, it is known from normal-state calculations that a finite spin magnetization is a necessary condition for SOC to produce dichroism. An alternative point of view is that, at $T = 0$, all paired electrons occupy mutually time conjugate states, so that the ground state is invariant under time reversal. Breaking this invariance, however, is mandatory for SOC-induced dichroism.

In Fig. 2 we keep the field fixed and vary the temperature. Above T_c the ratio is unity. Right below T_c we find a drastic increase in dichroism in the superconductor [9]. This increase can be traced back partly to a Hebel-Slichter-like effect, due to a novel combination of coherence factors. However, this mechanism alone cannot account for the size of the peak. A second amplifying mechanism is due to the presence of the superconducting gap. A spin-dependent gap in the band structure can strongly enhance SOC-induced dichroism because, in the presence of a magnetic field, transitions across the gap become energetically impossible for some quasiparticles (QP) of one spin direction, while they are still possible for the corresponding QP of the other spin. Because of the SOC, this difference between transitions of spin-up and spin-down QP results in a corresponding difference in the absorption of polarized light, in the same way as in the normal state [1]. Hence an additional increase in dichroism results from the gap [10]. Both mechanisms together, the Hebel-Slichter enhancement and the gap enhancement, account for the full peak in Fig. 2.

The second graph in Fig. 2 is for B close to Δ. The peak below T_c is not affected much, but at $T = 0$ the curve approaches a finite value. When the Zeeman energy

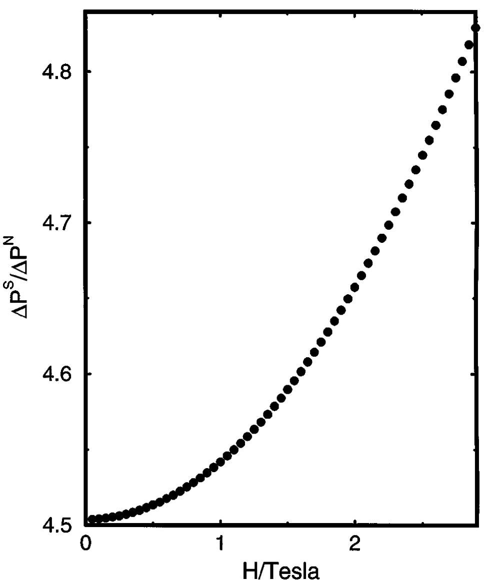

FIG. 1. Dichroism ratio vs magnetic field at $T = 3$ K for $\omega = 3$ meV, and an energy gap at $T = 0$ of 2 meV.

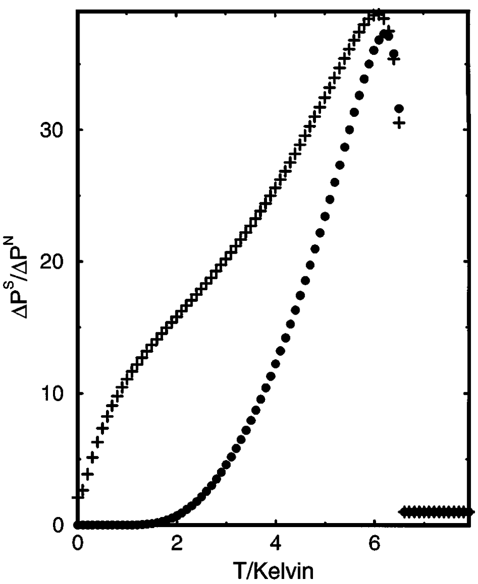

FIG. 2. Dichroism ratio vs temperature for $\omega = 4$ meV and $B = 0.1$ T (dots) and $B = 17.5$ T (crosses). T_c is at 6.6 K. Hebel-Slichter and gap enhancement are seen.

of the electron spins is comparable to the energy gap, the magnetic field can break Cooper pairs paramagnetically. Once there are unpaired electrons, the magnetic field immediately produces a net spin magnetization (hence, breaks time reversal invariance), and dichroism results. Interestingly, this effect could lead to a very direct experimental identification of paramagnetic limiting as the cause of the upper critical field.

In Fig. 3, finally, we display the frequency dependence of the absorption. There is an absorption edge at $\omega = 2$ meV, which corresponds to the energy gap. This edge occurs because we considered only pair breaking processes [11]. The behavior right above the edge is usually classified phenomenologically according to the shape of the peak to be type-I or type-II [4], a classification which reflects the symmetry under time reversal of the perturbation (type-I: odd; type-II: even). The shape of the peak in Fig. 3 is of a mixed type, reflecting the fact that our model contains two perturbations, the electron-photon interaction Hamiltonian (which is odd under time reversal) and the SOC (which is even).

The various plots demonstrate that even in the simple model many characteristics of the physics of dichroism and of superconductivity can be found. The quantitative details depend, of course, on the parameters of the model. However, we believe that the qualitative behavior is generic, and constitutes a definite signature of the SOC mechanism. For experimental investigations of the SOC mechanism we suggest Pb because its material parameters are similar to those used in our model calculations and its high atomic number of 82 implies strong spin-orbit coupling.

We end this Letter by noting that dichroism in the far-infrared absorption by $YBa_2Ca_3O_{7-\delta}$ films has been studied experimentally by Wu *et al.* [12] and Lihn *et al.* [13]. Our model calculations are not directly applicable to these experiments which, at low temperatures, appear to probe the orbital currents in the vortex state [12,13]. Evidently, to explain these, we would have to invoke our mechanism 3, which involves orbital currents. It follows from Eq. (7) that $\lim_{T\to 0} \mathrm{Im}[\hat{\sigma}_{xy}]$ is finite for the orbital mechanism, while, as demonstrated above, it is zero for the SOC mechanism. This supports the claim of Wu *et al.* that they are seeing cyclotron resonance, because their results do indeed extrapolate to a finite value at $T = 0$. Furthermore, the analysis of the experiments suggests that orbital currents alone do not provide a full explanation of the data and leads to the conclusion that there may be some missing “chiral resonances” [13] at work. Evidently, our spin-orbit effects, SOC and ASOC, could play the role of such resonances.

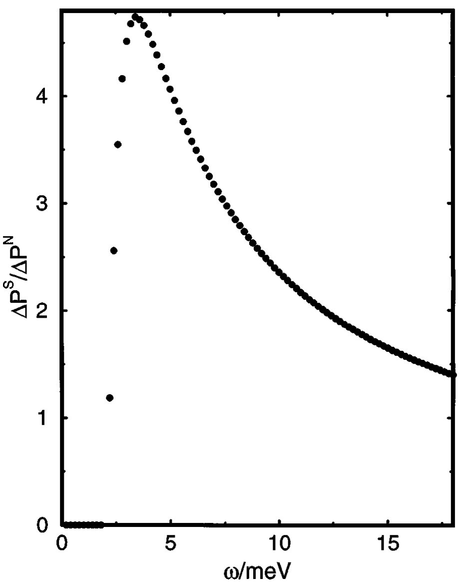

FIG. 3. Dichroism ratio vs frequency at $T = 3$ K and for $B = 0.1$ T. A mixed type-I–type-II absorption edge at the gap energy of 2 meV is present.

This work has benefited from collaborations within, and has been partially funded by, the HCM network, *Ab initio (from Electronic Structure) Calculation of Complex Processes in Materials (Contract No. ERBCHRXCT930369)*, and the program, *Relativistic Effects in Heavy-Element Chemistry and Physics*, of the European Science Foundation. Partial financial support by the Deutsche Forschungsgemeinschaft is gratefully acknowledged. We wish to thank H. Ebert, R. Kümmel, H. Plehn, and M. Lüders for many valuable discussions.

[1] *Spin-Orbit Influenced Spectroscopies of Magnetic Solids*, edited by H. Ebert and G. Schütz (Springer, Heidelberg, 1996); H. Bennett and E. Stern, Phys. Rev. **137**, A448 (1965); H. Ebert, Rep. Prog. Phys. **59**, 1665 (1996).
[2] P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).
[3] K. Capelle and E. K. U. Gross, Phys. Lett. A **198**, 261 (1995).
[4] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).
[5] K. Capelle, E. K. U. Gross, and B. L. Györffy (to be published).
[6] Q. P. Li and R. Joynt, Phys. Rev. B **44**, 4720 (1991); B. Janko and J. Shore, Phys. Rev. B **46**, 9270 (1992); Y. Zhu, F. Zhang, and H. Drew, Phys. Rev. B **47**, 586 (1993).
[7] L. C. Hebel and C. P. Slichter, Phys. Rev. **107**, 901 (1957).
[8] D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).
[9] Below T_c the normal-state calculation, strictly speaking, loses its meaning, but it still constitutes a convenient normalization, in particular, since the response of the *normal* conductor does not change much between $T = 0$ and $T = T_c$.
[10] A similar mechanism was also discussed for Heusler alloys, which also feature a spin-dependent gap [R. A. de Groot *et al.*, J. Appl. Phys. **55**, 2151 (1984)].
[11] Inclusion of single-particle scattering produces a (generally small) additional absorption below the edge [4].
[12] S. Wu *et al.*, Phys. Rev. B **54**, 13 343 (1996).
[13] H.-T. Lihn *et al.*, Phys. Rev. Lett. **76**, 3810 (1996).