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Author

Moore, D.

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Dexter Moore and Ignacio Tinoco, Jr.

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THE CIRCULAR DICHROISM OF LARGE HELICES. A FREE PARTICLE
ON A HELIX.

Dexter Moore
Chemistry Department
Temple University
Philadelphia, PA 19122

Ignacio Tinoco, Jr.
Chemistry Department and Laboratory of Chemical Biodynamics
University of California, Berkeley
Berkeley, CA 94720

ABSTRACT

Equations are derived for the circular dichroism and optical rotation of light by a free electron on a helix which is of arbitrary size relative to the wavelength of light. The equations were found to give results in good agreement with measured rotation of microwaves by oriented wire helices. The derivation shows when the usual Rosenfeld approximation for optical activity becomes inadequate.

In 1957 experimental work on the rotation of linearly polarized microwave radiation by copper wire helices was published.¹ The theoretical discussion of these results has been very tardy. A free particle on a helix small compared to the wavelength of light was discussed in 1964,² but the results were inapplicable to the microwave experiments. For helices large compared to the wavelength it is necessary to keep the vector potential of the light in the interaction Hamiltonian, instead of making the expansion which leads to the usual Rosenfeld³ approximation for optical rotation. The correct vector potential has been used earlier in treatments of optical rotation and circular dichroism of helices, but only to explicitly use the helical symmetry to obtain correct selection rules.⁴⁻⁶ Balazs et al.⁷ have derived a general expression for the average optical rotation of a free particle on a space curve of arbitrary size. However, they did not explicitly consider oriented helices. We will limit ourselves to large helices and in particular to oriented helices. The results are directly applicable to the microwave-wire helices experiments; they may also be used as guides for the interpretation of data obtained on biological helices as found in viruses, chromosomes and cells.⁸⁻¹⁰

The optical activity of a system of arbitrary size can be calculated from the following equations.¹¹

For light incident along unit vector $\underline{e}_1$ the circular dichroism (units are molar⁻¹cm⁻¹) is given by

$$\epsilon_L - \epsilon_R = \frac{24\pi^3 N_O \lambda g(\lambda)}{6909hc} \left(\frac{e\lambda_{OA}}{2\pi\mu c} \right)^2 \text{Im}[\underline{e}_1 \cdot \underline{T}_{AO}^* \times \underline{T}_{OA}] \quad (1)$$

and the optical rotation (units of degrees molar⁻¹cm⁻¹) is

$$[O] = \left(\frac{72\pi N_O}{hc} \right) \left(\frac{e\lambda_{OA}}{2\pi\mu c} \right)^2 \left(\frac{\lambda_{OA}^2}{\lambda^2 - \lambda_{OA}^2} \right) \text{Im}[\underline{e}_1 \cdot \underline{T}_{AO}^* \times \underline{T}_{OA}] \quad (2)$$

For light polarized along $\underline{e}$ the absorption is

$$\epsilon = \frac{24\pi^3 N_O \lambda g(\lambda)}{6909hc} \underline{e} \cdot \underline{T}_{OA} \underline{T}_{AO}^* \cdot \underline{e} \quad (3)$$

Here

$$\underline{T}_{OA} = \int \psi_O^* \exp(i2\pi \underline{e}_1 \cdot \underline{r} / \lambda_{OA}) \underline{p} \psi_A d\tau \quad (4)$$

and $\underline{r}$, $\underline{p}$ are position and linear momentum operators; e, μ are the charge and mass of an electron; c is the speed of light; λ is the wavelength of light and $g(\lambda)$ is a band shape function of unit area; N_O is Avogadro's number. ψ_O, ψ_A are the wavefunctions corresponding to energies E_O, E_A ; λ_{OA} is equal to $hc/(E_A - E_O)$. Evaluation of the matrix elements defined in Eq. (4) thus suffice to obtain the optical properties.

FREE ELECTRON ON A HELIX

The wavefunctions and operators for a free electron on a helix are given in reference 2. For a helix of k turns, radius a and pitch $2\pi b$ oriented along the z axis:

$$\psi_n = (k\pi)^{\frac{1}{2}} \sin(n\theta/2k)$$

$$x = a \cos \theta \quad y = a \sin \theta \quad z = b\theta$$

$$p_x = [i\hbar/(a^2+b^2)] [\sin \theta (\partial/\partial \theta) + \frac{1}{2}(\cos \theta)]$$

$$p_y = [-i\hbar/(a^2+b^2)] [\cos \theta (\partial/\partial \theta) - \frac{1}{2}(\sin \theta)]$$

$$p_z = [-i\hbar/(a^2+b^2)] (\partial/\partial \theta)$$

The selection rules for the matrix elements (Eq. 4) depend only on the direction of incidence of the light. For an integral number of turns, k , and quantum numbers m, n the selection rules are:

x incidence	$m-n$ odd
y incidence	$m-n$ odd, $m \pm n = 2k, 6k, 10k \dots$
z incidence	all transitions

The values of the non-zero matrix elements are:

x incidence, m-n odd

$$T^x(p_y) = -aC \sum_{r=-\infty}^{\infty} i^r J_r(a') [F_+ + F_- + i2a'k^2(E'_+ + E'_-)] \quad (5)$$

$$T^x(p_z) = -bC \sum_{r=-\infty}^{\infty} i^r J_r(a') [2F + i4a'k^2(E_+ + E_-)]$$

y incidence, m-n odd

$$T^y(p_x) = -iaC \sum_{r=-\infty}^{\infty} J_r(a') [F_+ + F_- + 2a'k^2(E'_+ + E'_-)] \quad (6)$$

$$T^y(p_z) = -bC \sum_{r=-\infty}^{\infty} J_r(a') [2F + 4a'k^2(E_+ - E_-)]$$

y incidence, m+n = ±2k(1+2r)

$$\begin{aligned} T^y(p_x) &= \left(\frac{-\pi aC}{8} \right) \left(\frac{m-n}{mn} \right) [J_{2r}(a') - J_{2r+2}(a')] \quad m+n=2k(1+2r) \\ &= \left(\frac{\pm \pi aC}{8} \right) \left(\frac{m+n}{mn} \right) [J_{2r}(a') - J_{2r+2}(a')] \quad m-n=\pm 2k(1+2r) \end{aligned} \quad (7)$$

$$\begin{aligned} T^y(p_z) &= \left(\frac{i\pi bC}{4} \right) \left(\frac{m-n}{mn} \right) J_{2r+1}(a') \quad m+n=2k(1+2r) \\ &= \left(\frac{\pm i\pi bC}{4} \right) \left(\frac{m+n}{mn} \right) J_{2r+1}(a') \quad m-n=\pm 2k(1+2r) \end{aligned}$$

z incidence

$$T^Z(p_x) = -iaC \left(\pm e^{i2\pi kb'} + 1 \right) \left(\frac{m^2 - n^2}{2} \right) [B_+ - B_-] \quad (8)$$

$$T^Z(p_y) = -aC \left(\pm e^{i2\pi kb'} + 1 \right) \left(\frac{m^2 - n^2}{2} \right) [B_+ + B_-]$$

Use + if (m-n) is odd; use - if (m-n) is even.

$$C = \frac{ihmn}{\pi k(a^2 + b^2)} \quad a' = 2\pi a / \lambda_{nm} \quad b' = 2\pi b / \lambda_{nm}$$

$$F_{\pm} = \frac{m^2 - n^2 - 4k^2 r(1 \pm r)}{(m^2 - n^2)^2 - 8k^2 (m^2 + n^2)(1 \pm r)^2 + 16k^4 (1 \pm r)^4}$$

$$F = \frac{m^2 - n^2 - 4kr^2}{(m^2 - n^2)^2 - 8k^2 (m^2 + n^2)r^2 + 16k^4 r^4}$$

$$E_{\pm} = \frac{(1 \pm r)}{(m^2 - n^2)^2 - 8k^2 (m^2 + n^2)(1 \pm r)^2 + 16k^4 (1 \pm r)^4}$$

$$E'_{\pm} = E_{\pm} \text{ with } (2 \pm r) \text{ replacing } (1 \pm r)$$

$$B_{\pm} = [(m^2 - n^2)^2 - 8k^2 (m^2 + n^2)(1 \pm b')^2 + 16k^4 (1 \pm b')^4]^{-1}$$

The components of the matrix elements are designated by the superscript which specifies the direction of incidence of the light and by the component of the linear momentum in parentheses. In evaluating the matrix elements the

special nature of the coordinate system requires that both possible orders of the exponential and momentum operators in Eq. (4) be used. The Bessel functions, $J_r(a')$, appear through the expansions

$$e^{ia'\sin\theta} = \sum_{r=-\infty}^{\infty} J_r(a') e^{ir\theta}$$

$$e^{ia'\cos\theta} = \sum_{r=-\infty}^{\infty} i^r J_r(a') e^{ir\theta}$$

In the limit of a' and b' approaching zero all the matrix elements (Eqs. 5-8) become equal to the matrix elements of the linear momentum operators.

OPTICAL ACTIVITY

To calculate the circular dichroism (Eq. 1) or the optical rotation (Eq. 2) one combines the appropriate components. For example, for z -incident light $\epsilon_L - \epsilon_R$ and $[\theta]$ are proportional to

$$\begin{aligned} \text{Im}[T^z(p_x)T^z(p_y)^* - T^z(p_x)^*T^z(p_y)] \\ = a^2 C^2 \left(\frac{m^2 - n^2}{2} \right)^2 (B_+^2 - B_-^2) (1 \pm \cos 2\pi k b') \end{aligned} \quad (9)$$

with (+) for $(m-n)$ odd and (-) for $(m-n)$ even. Similar equations apply for x - and y - incident light. The

calculated optical activity becomes equal in the appropriate limits to that published (see Eq. A2 in reference 2) in 1956 for the Rosenfeld approximation. The only term that requires comment is for z-incident light, where the denominator of $B_{\pm} = 0$ where $(m \pm n) = \pm [2k(1 \pm b')]$. In principle this can not happen exactly because $b' \equiv 2\pi b / \lambda_{nm}$ can never be an integer. However, in the limit of $b' \rightarrow 0$ the expressions for $(m-n) = \pm 2k$ or $(m+n) = 2k$ must be evaluated as special cases.

We are interested in the general trends that occur as the wavelength of light becomes equal or smaller than the helix dimensions. For our model the wavelength and helix size are not independent as

$$\lambda_{nm} = \frac{32\pi^2 \mu k^2 (a^2 + b^2) c}{h(m^2 - n^2)} \quad (10)$$

For x- and y- incident light (perpendicular to the helix axis) the important parameter is the radius of the helix, a . This determines how many, and which, Bessel functions contribute to the optical activity. As long as $(a' \equiv 2\pi a / \lambda_{nm}) \leq 0.2$ the Rosenfeld approximation is reasonable.¹¹ For light incident parallel to the helix axis the important parameter is the pitch, b , times the number of turns of the helix, k . As long as the parameter $(2\pi b' k \equiv 4\pi^2 b k / \lambda_{nm})$ is less than or equal to 0.2,

the Rosenfeld approximation is reasonable. The values of $b' \equiv 2\pi b/\lambda_{nm}$ also determine the zero's and extrema for the optical activity along the helix axis. Although b' cannot be an integer or rational fraction (see Eq. 10), as b' approaches 0, $1/k$, $2/k$, ... for $(m-n)$ even or $1/2k$, $3/2k$, $5/2k$ for $(m-n)$ odd, the optical activity for light incident along the helix axis approaches zero because of the cosine term in Eq. 9. This is true except for the special case when $m \pm n$ approximately equal $\pm[2k(1 \pm b')]$. Now both the numerator and denominator of Eq. (9) approach zero and a non-zero value for the optical activity results.

Some aspects of the optical activity are illustrated in Figures 1 and 2 where relative circular dichroism is plotted vs. a' or b' . In these figures the wavelength λ_{nm} is kept approximately constant by choosing $(m-n) = 1$ and allowing $(m+n)$ to vary as the helix parameters a and b change. For simplicity a one turn helix is chosen with $a = b$. In Fig. 1 the calculated contribution to the circular dichroism is compared with that calculated from the Rosenfeld approximation. One notices that the circular dichroism increases very rapidly as the size of the helix increases relative to the wavelength, and that the Rosenfeld approximation is useful even up to $a' = b' = 0.5$. Fig. 2 shows the contribution to the circular dichroism for one wavelength

and for light incident along the helix axis. Note that the circular dichroism approaches zero for $b' = 2.5, 3.5, 4.5, 5.5$ as expected for $(m-n)$ odd. For $b' = 0.5$ and 1.5 the circular dichroism is not close to zero, because these are the special cases of $(m-n) = \pm[2k(1 \pm b')]$.

It is interesting to note in Fig. 2 that the sign of the circular dichroism for this transition does not change as the size of the helix increases. The generalization deduced for the Rosenfeld approximation about the sign of the circular dichroism and the sense of a helix is valid for any wavelength. If a system is approximated by a free electron on a helix containing $2N$ electrons, the longest wavelength transition is from N to $N+1$ ($m-n = 1$). For a right handed helix this transition will always have a negative contribution to the circular dichroism for light incident parallel to the helix axis. It will have a positive contribution for light incident perpendicular to the helix axis.

LONG HELICES

In the limit as the number of turns of the helix, k , becomes very large the values of the matrix elements simplify. There are two classes of transitions which must be considered: $(m-n)$ of the same order as k and $(m-n)$ much less than k . When $(m-n)$ is much less than

k , λ_{nm} is proportional to k and a' and b' are small. This means the Rosenfeld approximation is valid; only $(m-n)$ odd transitions are allowed. The contribution to the circular dichroism for light incident along the helix axis approaches zero, and the circular dichroism for light incident along x is equal to that for light incident along y . For $(m-n)$ of the same order as k the selection rule is $(m-n)=2k(1+2r)$ with r an integer. The contributions of these transitions to the circular dichroism do depend on whether the light is incident along x or y . That is, although the helix is very long the absorption and the circular dichroism depend on whether the light is incident along x or y (both perpendicular to the helix axis). In fact there is negligible absorption or circular dichroism for light incident along x , but finite values for light incident along y or along the helix axis.

COMPARISON WITH EXPERIMENT

Measurements of the rotation of polarized microwaves by wire helices¹ provide the best application for these equations. The helices had a radius of 0.25 cm and a pitch 0.33 cm with three turns. The wavelength range studied was 2 to 4 cm, therefore the characteristic parameters were $a' = 0.4$ to 0.8 and $2\pi kb' = 1.5$ to 3 . The Rosenfeld approximation is obviously not applicable.

To apply the present equations to the wire helices it is convenient to rewrite them for the special case of large quantum numbers. We assume that there are N levels filled with electrons where N is much larger than k , the number of turns of the helix. Now m and n are both approximately equal to N and are much larger than k , but $(m-n)$ is of the same order as k . If we designate $(m-n)$ as j , a positive integer, then we can write the transition wavelength as approximately

$$\lambda_j = \frac{16\pi^2 \mu c k^2 (a^2 + b^2)}{h N j} = \frac{4 \mu c L^2}{h N j} \quad (11)$$

where $L = 2\pi k(a^2 + b^2)^{1/2}$ is the length of wire in the helix and c is the speed of light. There are j transitions that approximately have this wavelength; they are the transitions from $N \rightarrow N+j$, $N-1 \rightarrow N-1+j$, $\dots$, $N-j+1 \rightarrow N+1$. For each non-zero transition for $m \approx n \approx N$ we have:

x incidence, j odd

$$T^x(p_y) = -iaG \sum_{r=-\infty}^{\infty} i^r J_r(a') D_r[j^2 - 4k^2(1+r^2)] \quad (12)$$

$$T^x(p_z) = -ibG \sum_{r=-\infty}^{\infty} i^r J_r(a') (j^2 - 4k^2 r^2)^{-1}$$

y incidence, j odd

$$T^Y(p_x) = 8ak^2 G \sum_{r=-\infty}^{\infty} J_r(a') r D_r \quad (13)$$

$$T^Y(p_z) = -ibG \sum_{r=-\infty}^{\infty} J_r(a') (j^2 - 4k^2 r^2)^{-1}$$

y incidence, j = 2k(1+2r)

$$T^Y(p_x) = \frac{i\pi aG}{4j} [J_{2r}(a') - J_{2r+2}(a')] \quad (14)$$

$$T^Y(p_z) = \frac{\pi bG}{2j} J_{2r+1}(a')$$

z incidence, j odd or even

$$T^Z(p_x) = 4ak^2 b' G D_b (\pm e^{i2\pi k b'} + 1) \quad (15)$$

$$T^Z(p_y) = -\frac{iaG D_b}{2} [j^2 - 4k^2 (1+b'^2)] (\pm e^{i2\pi k b'} + 1)$$

Use + for j odd and - for j even

$$G = 8\mu ck/\lambda_j$$

$$D_r = [j^4 - 8k^2 j^2 (1+r^2) + 16k^4 (1-r^2)^2]^{-1}$$

D_b = same as D_r , but with b' substituted for r

These equations can be used to calculate the circular dichroism contribution of each of the j transitions at wavelength λ_j .

We have used Eq. (2) plus Eqs. (11-15) to calculate the rotation of polarized microwaves by oriented wire helices. Experimentally it was found that anomalous rotatory dispersion occurred at wavelengths equal to a constant divided by an integer, as expected from Eq. (11). These wavelengths occurred at about 3% shorter wavelength for light incident perpendicular to the helix axis than for light incident parallel to the helix axis; this does not follow from our equations. However, only odd integers corresponded to the anomalous dispersion for perpendicular incident light, while all integers led to anomalous dispersion for parallel incident light, as expected. The magnitude of the rotations had a wavelength dependence consistent with Eq. (2): a Drude dependence. The data were fit to a Drude curve and the coefficient corresponding to each anomalous dispersion region was obtained. To compare with our calculations we used the experimental values of λ_j for light incident parallel to the helix, and the helix dimensions: $a = 0.254$ cm, $b = 0.0530$ cm, $k = 3$. We calculated rotational strengths in $(\text{esu-cm})^2$ for each transition. In the Rosenfeld approximation the average rotational strength is $\text{Im } \underline{\mu}_{OA} \cdot \underline{m}_{AO}$; the correct expression for an oriented system is

$$\left(\frac{e\lambda_{OA}}{2\pi\mu c} \right)^2 \text{Im}[\underline{e}_1 \cdot \underline{T}_{AO}^* \times \underline{T}_{OA}]$$

The calculated values for five transitions are shown in Table 1. The values include the effect of the degeneracy; for example, as there are two transitions corresponding to λ_2 the rotational strength has been multiplied by two. For light incident parallel to the helix axis R_j^Z was evaluated exactly; for light incident perpendicular to the helix axis R_j^X, R_j^Y were obtained from the first three Bessel functions, $J_0, J_{\pm 1}, J_{\pm 2}$. The magnitudes of the rotational strengths are seen to correspond roughly to the motion of 0.1 electronic charge through 1 cm. It is of interest to note that values of λ_j correspond to quantum numbers N of order 10^{10} from Eq. (11). The calculated values of the rotational strengths are compared in Table I with the experimental Drude anomalous dispersion parameters. The rotational strength at $\lambda_4 = 2.36$ cm was normalized to the experimental value of -24.2 and the other rotational strengths were scaled by the same factor. The values of R_j^X and R_j^Y were averaged to obtain the perpendicular parameter. The agreement is excellent. The relative magnitudes of all the terms are correct, but most important is the fact that the zeros are correct. Use of the Rosenfeld approximation predicts the wrong magnitudes and incorrectly predicts zeros for R_j^Z for $j = 2$ and 4.

POLYMER AGGREGATES AND LIQUID CRYSTALS

Viruses, chromosomes, cholesteric liquid crystals and other large-scale aggregates of molecules show very large rotations of polarized light and very large apparent circular dichroism.⁸⁻¹⁰ The phenomena seem to correspond to almost complete reflection of one circularly polarized component and transmission of the other. The present model can be used to calculate the scattering of circularly polarized light and may give further insight into the very interesting optical properties of these systems.

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FIGURES

1. The contribution to the circular dichroism from one transition as a function of the ratio of the helix dimensions to wavelength. A one-turn right handed helix with $a=b$ is used. A transition for which $m-n = 1$ is chosen and λ_{nm} is kept constant by varying $m + n$ so that $(a^2 + b^2)/(m + n)$ is constant. The sign of the circular dichroism for y-incident light is positive and the sign for z-incident light (along the helix axis) is negative. The results are compared with the Rosenfeld approximation.
2. The contribution to the circular dichroism for light incident along the helix axis (z-incidence) for the transition and helix described in Figure 1. The sign of the circular dichroism is negative.

Table I. Calculated rotational strengths for the rotation of polarized microwaves by wire helices. The experimental data (from ref. 1) were obtained by fitting the measured rotations to a Drude equation. The value of the calculated rotational strength at $\lambda = 2.36$ cm was used to normalize the other rotational strengths to compare with experiment. The experimental perpendicular rotations are compared with the average of R^x and R^y . The theoretical values given here differ somewhat from earlier numbers presented at a NATO Conference in 1978 and published in "Optical Activity and Chiral Discrimination," S. F. Mason, editor, D. Reidel Publishing Co., 1979.

J	λ_j (cm)	Rotational Strengths			Relative Rotations			
		$\times 10^{22}$ esu ² cm ²			Parallel		Perpendicular	
		R^x	R^y	R^z	Expt.	Theory	Expt.	Theory
1	9.44	74.6	76.1	-0.2	-	- 0.2	+83	+56
2	4.72	0	0	-0.5	-	- 0.4	0	0
3	3.15	5.5	8.2	-1.6	- 2.2	- 1.2	+4.6	+5.1
4	2.36	0	0	-32.5	-24.2	(-24.2)	0	0
5	1.89	-19.3	2.7	-130	-106	-97	-	-6.2

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