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Notes on Optical Constants. By Mr C. G. DARWIN, F.R.S., Tait Professor of Natural Philosophy in the University of Edinburgh.

I. *The Optical Behaviour of certain Atomic Models.*

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§ 1. In a recent paper* the present writer made an attempt to reduce the ordinary optical constants of matter to a more primitive form by introducing "scattering indices." Their use solves half the problem of optics, for general formulae connect these indices with the behaviour of matter in bulk. There remains the deduction of the scattering indices from whatever we are assuming to be the reaction to light of the actual atoms, and even in the classical theory this encounters rather formidable difficulties of convergence. As long as we are content with no great rigour it is a fairly simple matter to deduce the scattering indices for any assumed model without any of those rather difficult arguments about the polarisation of the medium which play a part in the ordinary presentation.

The essential point which simplifies the process is that the wave given out by each atomic model is spherical and contains therefore a factor $e^{ik(ct-r)}/r$. So as long as we consider a point at a distance of only a small fraction of a wave-length away kr is small and the field is practically *statical*. This makes it easy to work out the Hertzian vector associated with the influence of an external electric force, and the factor e^{-ikr} can be re-introduced afterwards. Though there is perhaps a defect in formal rigour in proceeding in this way, there can I think be no doubt of its correctness, and it certainly leads to the right result in all cases where it can be compared with more precise methods.

We shall first illustrate the procedure with the model with which Lorentz obtains the ordinary dispersion formula, and shall then establish a result quoted but not proved in § 20*, which shows how the magnetic gyration of light can be connected with the idea that magnetism is equivalent to a rotation. Lastly, we shall investigate Drude's† "hypothesis of molecular currents." This hypothesis has I think been abandoned completely, but in the

* "The Optical Constants of Matter," *Camb. Phil. Soc. Trans.* vol. XXIII, No. VI (1924). I shall use the notation of the paper without explanation. References to formulae given there will be marked by a *.

† Drude, *Theory of Optics* (English Translation), p. 418.

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course of a study of the dispersion of magnetic gyration certain very peculiar experimental results† (with oxygen) suggested that it would be worth re-examining. The present method involves rather troublesome algebra but must I think be regarded as more rigorous than Drude's, and leads to quite different results. The two processes are too far apart to make it easy to see where the discrepancy arises, and I have not attempted to do so in detail, partly because the hypothesis has been abandoned and also because Drude works throughout his book on the inferior principle which obtains the Sellmeier and not the Lorentz dispersion formula.

In the course of the present work I have gained a good deal of experience with the tensor notation and of its utility. The verdict must I think be that as long as we are concerned with pure vectors it is inferior to the vector notation in quickness and convenience, but that the slightest degree of complication, as for instance double refraction, puts the vector notation into difficulty and brings out the power of the tensor algebra.

§ 2. Consider the atom as composed of an electron of charge e which can vibrate isotropically about the origin. Under the influence of an electric force $E_a{}^0 e^{ikct}$ its position ξ_a is given by

$$e\xi_a = \rho E_a{}^0 e^{ikct} \qquad \ldots\ldots(2{\cdot}1).$$

If the restoring force is $mk_0{}^2c^2$, we have

$$\rho = e^2/mc^2\,(k_0{}^2 - k^2) \qquad \ldots\ldots(2{\cdot}2),$$

but beyond noting that ρ is a function of k we shall not make any use of this relation. The rigorous deduction of the radiation from (2·1) is well known, but we shall give here a shorter non-rigorous process which will illustrate our method.

Consider a point x_a a small fraction of a wave-length distant from the electron. The electrostatic potential there is $\phi = e/r_1$, where $r_1{}^2 = (x_a - \xi_a)^2$. We suppose that ξ_a is much smaller than x_a and so have $\dfrac{1}{r_1} = \dfrac{1}{r} - \xi_a \dfrac{\partial}{\partial x_a}\dfrac{1}{r}$, where $r^2 = x_a{}^2$. Now if Π_a is the Hertzian vector $\phi = -\partial\Pi_a/\partial x_a$. Hence considering only the variable part we have $\Pi_a = e\xi_a/r = \rho E_a{}^0 e^{ikct}/r$. Π_a satisfies the wave equation, and to fulfil this condition at other distances we therefore introduce a factor e^{-ikr} and have

$$\Pi_a = \rho E_a{}^0 e^{ik(ct-r)}/r.$$

Let there be N such atoms in unit volume. Then the volume dV gives rise to a scattered wave of amount

$$\Pi_a = N dV \rho E_a{}^0 e^{ik(ct-r)}/r.$$

This is equivalent to (4·1)*

$$\Pi_a = (\sigma/4\pi)\, dV E_a{}^0 e^{ik(ct-r)}/r.$$

† Siertsema, *Arch. Neerl.* vol. II, p. 291 (1899).

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Thus $\sigma = 4\pi N\rho$ and we obtain exactly Lorentz's dispersion by introducing (2·2).

§ 3. We can apply the same method to connect the magnetic gyration of light with the Larmor rotation. An atom in a magnetic field $H = \frac{2mc}{e}\omega$ behaves as though there were no field provided it is regarded from the point of view of axes rotating with angular velocity ω in the negative sense. We suppose the atom isotropic and that its vibration is described by the coefficient ρ of § 2, but we must now remember that ρ is a function of k. It is exposed to an electric force $E_1{}^0 e^{ikct}$. If this is referred to axes rotating with velocity $-\omega$ about x_3 we have

$$E_1' = E_1{}^0 e^{ikct}\cos\omega t = \tfrac{1}{2}E_1{}^0 e^{i(kc+\omega)t} + \tfrac{1}{2}E_1{}^0 e^{i(kc-\omega)t},$$
$$E_2' = E_1{}^0 e^{ikct}\sin\omega t = -\tfrac{1}{2}iE_1{}^0 e^{i(kc+\omega)t} + \tfrac{1}{2}iE_1{}^0 e^{i(kc-\omega)t}.$$

Referred to these axes the atom is isotropic; so in its neighbourhood we shall have

$$\Pi_1' = \tfrac{1}{2}\rho\left(k+\frac{\omega}{c}\right)E_1{}^0 e^{i(kc+\omega)t}/r + \tfrac{1}{2}\rho\left(k-\frac{\omega}{c}\right)E_1{}^0 e^{i(kc-\omega)t}/r,$$

$$\Pi_2' = -\tfrac{1}{2}i\rho\left(k+\frac{\omega}{c}\right)E_1{}^0 e^{i(kc+\omega)t}/r + \tfrac{1}{2}i\rho\left(k-\frac{\omega}{c}\right)E_1{}^0 e^{i(kc-\omega)t}/r.$$

Transform these back to fixed axes, and we have

$$\Pi_1 = \tfrac{1}{2}\left\{\rho\left(k+\frac{\omega}{c}\right)+\rho\left(k-\frac{\omega}{c}\right)\right\}E_1{}^0 e^{ikct}/r,$$

$$\Pi_2 = -\frac{i}{2}\left\{\rho\left(k+\frac{\omega}{c}\right)-\rho\left(k-\frac{\omega}{c}\right)\right\}E_1{}^0 e^{ikct}/r.$$

To describe the effects at greater distances we now introduce the factor e^{-ikr}. If the incident light is not very close to a region of resonance we can expand ρ and have

$$\Pi_1 = \rho E_1{}^0 e^{ik(ct-r)}/r,$$

$$\Pi_2 = -i\frac{\omega}{c}\frac{d\rho}{dk}E_1{}^0 e^{ik(ct-r)}/r.$$

Summing over unit volume we obtain as the scattering indices

$$\sigma_{11} = 4\pi N\rho,$$

$$\sigma_{12} = -4\pi N i\frac{\omega}{c}\frac{d\rho}{dk},$$

where the σ's are defined as in (11·2)*, and so we have proved

$$\sigma_{12} = -i\frac{\omega}{c}\frac{d\sigma}{dk}.$$

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This result does not in any way depend on the form of σ, and so will be true, for example, for the Kerr effect, provided that it is legitimate there to regard the magnetism as equivalent to a rotation.

§ 4. In Drude's "hypothesis of molecular currents" an electron is supposed to describe a small circle rapidly, and under the influence of the electric field the whole circle shifts to and fro. This has the effect of loading the magnetic flux in the medium. The model is equivalent to supposing that to each electron is attached a magneton. In an external magnetic field the magnetons are orientated, and as they oscillate they will cause an emission. We shall work out the scattering of such a system.

Take a magneton of which the components have moment $e\mu_\theta$. (The factor e, the charge of the attached electron, is introduced for later simplification; μ_θ is of the physical dimensions of a length.) The vector potential of the magneton is

$$A_\beta = [\beta\gamma\theta]\, e\mu_\theta \frac{\partial}{\partial x_\gamma}\left(\frac{1}{r_1}\right) \qquad \text{.....(4·1).}$$

As in § 2 we replace $\frac{1}{r_1}$ by $\frac{1}{r} - \xi_\delta \frac{\partial}{\partial x_\delta}\frac{1}{r}$, and drop the constant part. We also substitute

$$e\xi_\delta = \rho E_\delta{}^0 e^{ikct} = \rho E_\alpha{}^0 (\alpha\delta)\, e^{ikct},$$

where we have introduced a "spherical tensor" $(\alpha\delta)$ so as to bring the expression into a standard form. We also have the relation

$$A_\beta = \frac{1}{c}\frac{\partial \Pi_\beta}{\partial t} = ik\Pi_\beta.$$

Hence

$$\Pi_\beta = -[\beta\gamma\theta]\,(\alpha\delta)\frac{\rho\mu_\theta}{ik} E_\alpha{}^0 \frac{\partial^2}{\partial x_\gamma \partial x_\delta}\frac{e^{ik(ct-r)}}{r} \qquad \text{.....(4·2).}$$

This looks as though it were of the standard form (11·2)*, but is not so as it does not conform to the "duplicate rule," § 12*, and the rule of symmetry between γ and δ. We can satisfy this last rule at once by permuting γ, δ and halving the result. Then (with R for $e^{ik(ct-r)}/r$)

$$\Pi_\beta = \frac{i\rho\mu_\theta}{2k}\{[\beta\gamma\theta]\,(\alpha\delta) + [\beta\delta\theta]\,(\alpha\gamma)\} E_\alpha{}^0 \frac{\partial^2}{\partial x_\gamma \partial x_\delta} R \quad \text{...(4·3).}$$

The application of the duplicate rule is more troublesome. We may recall that there are no waves of the second rank associated with $\sigma_{\alpha\beta\beta\delta}$, $\sigma_{\alpha\beta\gamma\beta}$, and that the wave associated with $\sigma_{\alpha\beta\gamma\gamma}$ is of zero and not the second rank. To satisfy the conditions take

$$\rho_{\alpha\beta\gamma\delta} = (i\rho\mu_\theta/2k)\{[\beta\gamma\theta]\,(\alpha\delta) + [\beta\delta\theta]\,(\alpha\gamma)\} + \rho'_{\alpha\beta}\,(\gamma\delta) + \rho''_{\alpha\gamma}\,(\beta\delta) + \rho''_{\alpha\delta}\,(\beta\gamma)$$

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and choose $\rho'_{\alpha\beta}$, $\rho''_{\alpha\gamma}$ so that $\rho_{\alpha\beta\gamma\delta}$ shall conform to the rule. To do this multiply by $(\beta\gamma)$. Observe that whereas $(\beta\gamma) \times (\gamma\delta)$ means that β is replaced by δ, we have $(\beta\gamma) \times (\beta\gamma) = 3$. Then

$$\rho_{\alpha\beta\beta\delta} = 0 = (i\rho\mu_\theta/2k)\,[\alpha\delta\theta] + \rho'_{\alpha\delta} + \rho''_{\alpha\delta} + 3\rho''_{\alpha\delta}.$$

Again multiplying by $(\gamma\delta)$

$$\rho_{\alpha\beta\gamma\gamma} = 0 = (i\rho\mu_\theta/2k)\,2\,[\beta\alpha\theta] + 3\rho'_{\alpha\beta} + \rho''_{\alpha\beta} + \rho''_{\alpha\beta}.$$

Changing δ into β we can solve and find

$$\rho'_{\alpha\beta} = 2\,(i\rho\mu_\theta/4k)\,[\alpha\beta\theta],$$
$$\rho''_{\alpha\delta} = -\,(i\rho\mu_\theta/4k)\,[\alpha\delta\theta] \qquad \ldots\ldots(4{\cdot}4).$$

The addition to (4·3) of $\rho''_{\alpha\delta}\,(\beta\gamma)$, $\rho''_{\alpha\gamma}\,(\beta\delta)$ gives no wave, but for $\rho'_{\alpha\beta}\,(\gamma\delta)$ this is not so, and we must allow for a compensating wave of zero rank. Thus

$$\Pi_\beta = (i\rho\mu_\theta/4k)\,E_\alpha\{2\,[\beta\gamma\theta]\,(\alpha\delta) + 2\,[\beta\delta\theta]\,(\alpha\gamma) + 2\,[\alpha\beta\theta]\,(\gamma\delta) - [\alpha\delta\theta]\,(\beta\gamma) - [\alpha\gamma\theta]\,(\beta\delta)\}\frac{\partial^2}{\partial x_\gamma \partial x_\delta} R$$
$$- (i\rho\mu_\theta/2k)\,E_\alpha{}^0\,[\alpha\beta\theta]\,(\gamma\delta)\,\frac{\partial^2}{\partial x_\gamma \partial x_\delta} R.$$

In the last line we can replace $(\gamma\delta)\frac{\partial^2}{\partial x_\gamma \partial x_\delta}$ by $-k^2$, which exhibits the reduction to zero rank.

We now add on the contribution from the electron as in § 2 and sum over the atoms in unit volume. The result is then exactly of the standard form in (11·2)* and we have

$$\sigma_{\alpha\beta} = \sigma\,(\alpha\beta) + \tfrac{1}{2}ik\sigma\mu_\theta\,[\alpha\beta\theta],$$
$$\sigma_{\alpha\beta\gamma\delta} = \tfrac{1}{4}ik\sigma\mu_\theta\{2\,[\beta\gamma\theta]\,(\alpha\delta) + 2\,[\beta\delta\theta]\,(\alpha\gamma) + 2\,[\alpha\beta\theta]\,(\gamma\delta) - [\alpha\delta\theta]\,(\beta\gamma) - [\alpha\gamma\theta]\,(\beta\delta)\}\ldots(4{\cdot}5).$$

This is the complete formal account of the optics of the medium. We observe that it differs essentially from the model of § 3 in that it contains a wave of the second rank. This suggests at once that the behaviour of the models will be different for magnetic double refraction, if they are adjusted to be the same for gyration.

§ 5. The equation of wave velocities is from (16·1)*

$$N_\eta = N_\alpha\left\{\left(\frac{1}{3} + \frac{1}{n^2-1}\right)\sigma_{\alpha\eta} - \frac{n_\eta n_\beta}{n^2-1}\sigma_{\alpha\beta} - \frac{n_\gamma n_\delta}{7}\,(\sigma_{\alpha\eta\gamma\delta} + \sigma_{\alpha\gamma\eta\delta} + \sigma_{\alpha\gamma\delta\eta}) - \frac{n_\gamma n_\delta}{n^2-1}\,(\sigma_{\alpha\eta\gamma\delta} - n_\eta n_\beta \sigma_{\alpha\beta\gamma\delta})\right\}\ldots(5{\cdot}1).$$

If we substitute from (4·5) only a few of the fourth rank terms survive; in particular the terms multiplied by 1/7 go right out,

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which shows that the convergence of the integrations is no more critical than in the ordinary type of medium. The equation reduces to

$$\frac{1}{\sigma}N_\eta = N_\alpha\left[\left(\frac{1}{3}+\frac{1}{n^2-1}\right)(\alpha\eta) - \frac{n_\eta n_\alpha}{n^2-1} - ik\mu_\theta\left\{\tfrac{1}{3}[\alpha\eta\theta] + \frac{n_\alpha n_\gamma}{n^2-1}[\eta\gamma\theta]\right\}\right] \quad \text{.....(5·2).}$$

To find the gyration we take magnetons $\mu_1 = \mu_2 = 0$, $\mu_3 = \mu$ and $n_1 = n_2 = 0$, $n_3 = n$. The equations become

$$\frac{1}{\sigma}N_1 = N_1\left(\frac{1}{3}+\frac{1}{n^2-1}\right) + N_2\frac{ik\mu}{3} \quad \text{.....(5·3),}$$

$$\frac{1}{\sigma}N_2 = N_1\left(-\frac{ik\mu}{3}\right) + N_2\left(\frac{1}{3}+\frac{1}{n^2-1}\right),$$

$N_3 = 0$. Thus

$$\frac{1}{3}+\frac{1}{n^2-1} = \frac{1}{\sigma}\pm\frac{k\mu}{3} \quad \text{.....(5·4),}$$

and

$$N_2 = \pm iN_1.$$

The gyration in unit length is given by $\frac{1}{2}k(n'-n'')$, where n' and n'' are the roots of (5·4). We have approximately

$$\tfrac{1}{2}k(n'-n'') = \frac{(n^2-1)^2}{2n}\frac{k^2\mu}{3}.$$

It is more interesting to compare it with the corresponding effect from § 3 where there is only a wave of zero rank. Instead of (5·4) we have in that case (20·3)*,

$$\frac{1}{3}+\frac{1}{n^2-1} = \frac{1}{\sigma\pm\tau},$$

where $\sigma_{12} = -\sigma_{21} = i\tau$. We thus have

$$\frac{\tau}{\sigma^2} = \frac{k\mu}{3} \quad \text{.....(5·5).}$$

For values of k rather near an absorption line σ varies as $1/(k_0^2-k^2)$, and τ by §.3 as $k/(k_0^2-k^2)^2$. Thus (5·5) indicates that Drude's model should give exactly the same dispersion as the other. For a wave length very close to the line the approximation fails, but it is easy to see that the gyration will not change sign. We thus conclude, in complete disagreement with Drude, that there will be nothing in optical observation of gyration to discriminate between the hypothesis of molecular currents and the hypothesis of rotation.

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§ 6. To study the double refraction we again take $\mu_1 = \mu_2 = 0$, $\mu_3 = \mu$, but now $n_1 = n$, $n_2 = n_3 = 0$. The equations are

$$\frac{1}{\sigma}N_1 = N_1\left(-\frac{2}{3}\right) + N_2\frac{ik\mu}{3} \qquad \text{.....(6·1),}$$

$$\frac{1}{\sigma}N_2 = N_1\, ik\mu\left(-\frac{1}{3} + \frac{n^2}{n^2-1}\right) + N_2\left(\frac{1}{3} + \frac{1}{n^2-1}\right),$$

$$\frac{1}{\sigma}N_3 = N_3\left(\frac{1}{3} + \frac{1}{n^2-1}\right).$$

The x_3 component is normal. For the other we have approximately

$$\frac{1}{3} + \frac{1}{n^2-1} = \frac{1}{\sigma} + \frac{1}{3}k^2\mu^2\frac{3+\sigma}{3+2\sigma} \qquad \text{.....(6·2)}$$

and

$$N_1 = -\frac{ik\mu\sigma}{3+2\sigma}N_2.$$

On the rotational hypothesis we have for the same case

$$\frac{1}{3} + \frac{1}{n^2-1} = \frac{1}{\sigma} + \frac{\tau^2}{\sigma^2}\frac{2}{3+2\sigma}.$$

If we substitute from (5·5) we make the gyrations the same, and the double refraction will then be in the ratio $3(3+\sigma) : 2\sigma^2$. We thus see that the double refraction will give a discrimination between the hypotheses, and where it has been observed it does of course in fact support the rotational. Drude's work again disagrees with ours, for he found no double refraction at all.

It appears to me that the present calculations start from exactly the same hypotheses as Drude's, and it is a little disquieting that they should give such different results, but the processes are too far apart to trace the discrepancy. Drude's neglect of the factor $\frac{1}{3}$ in calculating refraction leads him to the Sellmeier formula instead of Lorentz's, and it is not unreasonable to suppose that this should make a capital difference in what is in essence a superposed second order effect. As the hypothesis has, I believe, been completely abandoned it does not seem worth while to search more deeply for the cause of the discrepancy.

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