Physics. — *On the* LORENTZ-LORENZ *Correction in Metallic Conductors.* By R. DE L. KRONIG and H. J. GROENEWOLD. (Communicated by Prof. H. A. KRAMERS).

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According to the theory of dispersion the index of refraction n of an isotropic substance for an electromagnetic wave of frequency ν is determined by the relation between the electric polarisation per unit volume P due to the wave and the macroscopic electric field E of the wave, entering into MAXWELL's equations, n being given by

$$n^2 - 1 = \frac{4\pi P}{E} \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (1)$$

In order to apply this formula it is necessary to know the relationship between P and E.

In the case of an insulator the customary procedure to obtain P/E is the following. For a particular atom of the substance the electric moment induced by the wave will be

$$p = \alpha E_0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (2)$$

where E_0 is the electric field of frequency ν acting on the atom and α the polarisability of the atom at that frequency. If we take as atom the harmonic oscillator usually employed in classical dispersion theory, i.e. an electron of charge $-e$ and mass m attached elastically to the electrical center of a fixed constellation of positive electricity with an equal total charge, then α is given by

$$\alpha = \frac{e^2}{4\pi^2 m (\nu_0^2 - \nu^2)}, \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (3)$$

ν_0 being the natural frequency of vibration of the electron. E_0 may in general not be identified with E. It can most easily be calculated by imagining a sphere constructed around the center of the atom under consideration, the radius being chosen small compared to the wavelength of the radiation but still so large that the sphere contains many atoms. Then

$$E_0 = E + E' + E'', \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (4)$$

where E' and E'' denote respectively the electric fields produced at the center of the atom considered by the polarisation of the atoms with centers

outside the sphere and by the polarisation of the other atoms with centers inside the sphere [1]). As LORENTZ [2]) showed, E' for an isotropic substance is given by

$$E' = \frac{4\pi}{3} P \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (5)$$

In order to compute E'', the arrangement of the atoms has to be known. For a simple cubical arrangement LORENTZ [3]) could prove that

$$E'' = 0 \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (6)$$

The same holds rigorously for a gas composed of independent atoms that can approach each other only up to a minimum distance [4]) and probably approximately for most isotropic substances. By combining eqs. (2), (4), (5) and (6) we get

$$P = Np = N\alpha\left(E + \frac{4\pi}{3} P\right),$$

where N is the number of atoms per unit volume. Solving for P/E and substituting in eq. (1) leads to the CLAUSIUS-MOSOTTI equation

$$3\frac{n^2 - 1}{n^2 + 2} = 4\pi N\alpha. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (7)$$

This equation differs from the equation

$$n^2 - 1 = 4\pi N\alpha. \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad . \quad (8)$$

which we would have obtained if we had neglected the LORENTZ-LORENZ correction, i.e. if we had identified E with E_0. Only when $4\pi N\alpha$ is small compared to unity is the difference between eqs. (7) and (8) inappreciable.

The question naturally arises as to how these considerations are altered when we are dealing with a metallic conductor. In order to illustrate the essential points we shall use as model of such a conductor a positive fluid of uniform charge density in which the electrons are situated at the corner points of a simple cubical lattice with a lattice constant a, so chosen that on the average the conductor is neutral. On the one hand it would seem as if the index of refraction ought now to be calculated in the following way: For a free electron in an electric field E_0 oscillating with frequency

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[1]) H. A. LORENTZ, The Theory of Electrons, Teubner 1909; p. 137 et seq.
[2]) H. A. LORENTZ, l. c.; p. 303 et seq.
[3]) H. A. LORENTZ, l. c.; p. 306.
[4]) R. LUNDBLAD, Ann. d. Phys. **57**. 183, 1918.

ν the product of its charge and its displacement from the original position of rest has the form

$$p = \beta E_0$$

with

$$\beta = -\frac{e^2}{4\pi^2 m\nu^2},$$

quite analogous to the relation (2) for an electron bound in an atom, so that replacing α by β in the CLAUSIUS-MOSOTTI equation (7) will give us n. On the other hand one can argue that no LORENTZ-LORENZ correction should be made in this case, or, expressing it differently, that eq. (8) with β instead of α should be applied, since all the electrons inside a region of linear dimensions small compared with the wave length suffer the same displacement, with the consequence that there are no electric forces of frequency ν acting on the electrons excepting the field E of the wave itself. In trying to decide between these two viewpoints it must be remembered that in the derivation of LORENTZ previously described it is essential that the substance be regarded as composed of neutral atoms in which the electrons are bound. By an artifice it is possible to look at our model of a metallic conductor in the same way, and it then appears that the force E', giving rise to the LORENTZ-LORENZ correction, just balances the elastic restoring force. In treating the electrons as free, the LORENTZ-LORENZ correction hence has already been taken into account so that the second method of calculating n proposed above is the correct one[1]).

We may imagine the positive fluid of our model subdivided into equal cubes by three mutually perpendicular sets of parallel planes. In particular we can chose these planes in such a way that in every cube there is one electron in the center. The length of the edge of the cubes will then be equal to the lattice constant a of the cubical lattice at the corner points of which we assumed the electrons to be located. If now, keeping the centers of the cubes, i.e. the positions of the electrons, fixed while letting the size of the cubes diminish (always retaining the positive charge with uniform density inside the cubes), we get a cubical lattice of "atoms" separated by finite intervals without any charge. The individual "atom", a cube filled with positive charge of uniform density and with an electron at its center, is such that we may apply eq. (3). Indeed, as will be shown in the note at the end of the paper, the electron for small displacements suffers an elastic

[1]) Prof. DARWIN first suggested to one of us in connection with a paper on the quantum theory of dispersion in metallic conductors (R. DE L. KRONIG, Proc. Roy. Soc. A. **124**, 409, 1929; see especially the footnoote on p. 419) that n should be computed according to the first method. The discrepancy between theory and experiment resulting in this way immediately led to serious doubts regarding this suggestion and to a more thorough investigation of the whole problem. The choice of the model employed here to elucidate the relation to the case of an insulator is the result of a discussion with Prof. KRAMERS.

restoring force directed toward the center of its cube, the natural frequency of vibration being given by

$$4\pi^2 m\nu_0^2 = \frac{a^3}{b^3}\frac{4\pi Ne^2}{3}, \quad \ldots\ldots\ldots \quad (9)$$

where b is the edge of the cube reduced in size as described above, while the other symbols have their old meaning. On the basis of what has been said in the beginning of this paper we may apply to our model the equation (7), obtaining

$$3\frac{n^2-1}{n^2+2} = \frac{4\pi Ne^2}{\frac{a^3}{b^3}\frac{4\pi Ne^2}{3} - 4\pi^2 m\nu^2}.$$

Now if b becomes equal to a, i.e. if the positive cubes touch, forming one continuous positive fluid, then this reduces to

$$n^2 - 1 = 4\pi N \cdot \left(-\frac{e^2}{4\pi^2 m\nu^2}\right) = 4\pi N\beta.$$

We can summarize our result as follows: *The index of refraction of our simple model of a metallic conductor may be calculated as if the electrons were free while the* LORENTZ-LORENZ *correction is omitted.*

There is in this argument still one point requiring proof, viz. that the value of ν_0, given for the isolated "atom" by eq. (9), is not altered by the close proximity of the neighbouring atoms. The proof is given in the note at the end. One will also inquire how the foregoing considerations are to be modified when the electrons combine the properties both of free and bound electrons as they do in the quantum theoretical treatment of metallic conduction developed by BLOCH. A discussion of this question will be reserved for a later investigation.

Note. We imagine a charge distribution in space, symmetrical with respect to the three coordinate planes, the density ϱ being continuous at the origin and having there the value ϱ_0. We wish to determine the electric field in the neighbourhood of the origin.

Let ξ, η, ζ be the coordinates of a point in the charge distribution, x, y, z the coordinates of a point near the origin, where the field is to be determined. If r denotes the length of the line joining the two points, then the x-component of the electric field due to the charge distribution is given by

$$E_x = -\frac{\partial V}{\partial x},$$

where

$$V = \int \frac{\varrho(\xi, \eta, \zeta)}{r} d\xi\, d\eta\, d\zeta.$$

For small values of x, y, z we may develop E in a power series in x, y, z, obtaining

$$E_x = -\left(\frac{\partial V}{\partial x}\right)_0 - \left(\frac{\partial^2 V}{\partial x^2}\right)_0 x - \left(\frac{\partial^2 V}{\partial x\,\partial y}\right)_0 y - \left(\frac{\partial^2 V}{\partial x\,\partial z}\right)_0 z,$$

the derivatives to be taken at the origin. On account of the symmetry of the charge distribution E_x vanishes at the origin. For the same reason the coefficients of y and z vanish. We retain

$$E_x = -\left(\frac{\partial^2 V}{\partial x^2}\right)_0 x = -x \cdot \int \frac{\varrho \cdot (3\xi^2 - r_0^2)}{r_0^5}\, d\xi\, d\eta\, d\zeta =$$

$$= -x \cdot \int \frac{\varrho}{3} \triangle\left(\frac{1}{r_0}\right) d\xi\, d\eta\, d\zeta,$$

r_0 being the value of r when x, y and z are zero. Taking a little sphere around the origin and splitting the integral into the contributions from outside and from inside this sphere, we get naught for the first and $-4\pi\varrho_0/3$ for the second part, where ϱ_0 is the charge density at the origin. We thus have

$$E_x = \frac{4\pi\varrho_0}{3} x$$

for small displacements from the origin. In other words an electron will be elastically bound to the origin, the frequency of vibration being given by

$$4\pi^2 m \nu_0^2 = \frac{4\pi\varrho_0 e}{3} \quad \ldots\ldots\ldots \quad (10)$$

In our model of a system of electrons situated at the points of a simple cubical lattice with lattice constant a, each electron being at the center of a cube with edge b, in which the positive charge e is uniformly distributed

$$\varrho_0 = \frac{a^3}{b^3} Ne.$$

Substitution in eq. (10) gives us eq. (9), and we also see now that the value ν_0 is the same whether we regard an isolated "atom" or an "atom" symmetrically surrounded by neighbours as in our model.

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