

FIG. 2

for heavy nuclei. Inasmuch as the  $C^{12}$  nucleus is not considered to be deformed, we have no relation between  $r_0$  and  $R_0$ , in contrast to the  $Mg^{24}$  case. Therefore we use the rough values  $R_0 = 4 \times 10^{-13}$  cm and  $r_0 = 6 \times 10^{-13}$  cm. In such a case,  $C = 24.4$  Mev.

If we assume here too that the principal maximum is connected with the nuclear interaction, we obtain  $V_0 = 5.41$  Mev. For the values of the parameters that we have chosen, it is seen that the electrical interaction plays almost no role in the angular distribution. The angular distribution obtained on the basis of Eq. (16) is shown by the solid curve in Fig. 2. As we see, the theoretical curve does

not have a minimum and disagrees, in many respects, with the experimental data. It agrees with experiment only in relation to the presence of the principal maximum.

It is possible that this nonconformity is produced by our incorrect assumption that, in the process considered by us, a single phonon excitation arises in the  $C^{12}$  nucleus. Nor is it excluded that, in such light nuclei as  $C^{12}$ , the generalized model is generally non-applicable.

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## DISPERSION OF LIGHT IN THE EXCITON ABSORPTION REGION OF CRYSTALS

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The theory of light waves in exciton-absorbing crystals, developed in Ref. 1 on the basis of a new relation between specific polarization and the electric field, is applied to cubic crystals. For each direction of propagation, the existence of three types of light waves is predicted in these crystals. One of these types is similar to ordinary waves, whereas the other two are essentially anomalous. The frequency dependence of the refractive indices of the three types is considered. Fresnel's formulas are generalized for light passing through the boundary between the crystal and a vacuum. New formulas are obtained for the coefficient of reflection from the crystal surface and for the transparency of a plane parallel plate. Methods are suggested for experimental testing of the theory and for obtaining a direct proof of the existence of second and third light in cubic crystals.

THE present article is an immediate continuation of Ref. 1, in which it was shown that in the region of exciton absorption of light the specific dipole

moment of dielectric polarization and the electric field are related through a differential equation rather than by a simple linear algebraic expres-

sion. Therefore Maxwell's equations are of a higher order and possess more than the customary number of solutions. Specifically it was shown in Ref. 1 that in a crystal there can exist several plane monochromatic waves of the same frequency, direction of propagation, and polarization but with different refractive indices (different velocities of propagation). This phenomenon differs from ordinary birefringence, where the waves of different refractive indices must be polarized at right angles to each other.

The next step to be taken is the extension of all equations of theoretical crystal optics to the excitation absorption region and the prediction of new effects which could be observed experimentally and thus provide a test of the theory. It is desirable to begin with cubic crystals in order to avoid the complication of these new effects by ordinary birefringence. We shall therefore apply here the theory of Ref. 1 to cubic crystals and predict several effects which are capable of experimental verification.

## 1. REFRACTIVE INDICES OF LIGHT

In Ref. 1 it was shown that when the solution of Maxwell's equations is sought with the electric and magnetic fields proportional to  $\exp\{i\omega[n(\mathbf{s}\cdot\mathbf{r})/c - t]\}$  for any given  $\omega$  and  $\mathbf{s}$ , three types of solution are possible in a cubic crystal:

$$\mathbf{E} = \mathbf{E}_+ \exp\left\{i\omega\left[\frac{n_+}{c}(\mathbf{s}\cdot\mathbf{r}) - t\right]\right\}, \quad \mathbf{E}_+ \perp \mathbf{s}; \quad (1)$$

$$\mathbf{E} = \mathbf{E}_- \exp\left\{i\omega\left[\frac{n_-}{c}(\mathbf{s}\cdot\mathbf{r}) - t\right]\right\}, \quad \mathbf{E}_- \perp \mathbf{s}; \quad (2)$$

$$\mathbf{E} = \mathbf{E}_\parallel \exp\left\{i\omega\left[\frac{n_\parallel}{c}(\mathbf{s}\cdot\mathbf{r}) - t\right]\right\}, \quad \mathbf{E}_\parallel \parallel \mathbf{s}. \quad (3)$$

The following expressions are obtained for the refractive indices:

$$n_\pm^2 = 1/2(\mu + \vartheta) \pm \sqrt{1/4(\mu - \vartheta)^2 + b}, \quad (4)$$

$$n_\parallel^2 = \mu', \quad (5)$$

with

$$\mu \equiv (2Mc^2/\hbar\omega)(1 - \mathcal{E}_0/\hbar\omega), \quad \mu' \equiv \frac{2M'c^2}{\hbar\omega}\left(1 - \frac{\mathcal{E}_0'}{\hbar\omega}\right), \quad (6)$$

where  $c$  is the velocity of light in a vacuum, and  $M$  and  $M'$  are the effective mass of an exciton for transverse and longitudinal polarization, respectively, which can be determined if the expansion of the energy of the exciton state in powers of the wave vector  $\mathbf{k}$  has the form

$$\mathcal{E}(\mathbf{k}) = \mathcal{E}_0 + \hbar^2\mathbf{k}^2/2M + \dots \quad \mathcal{E}'(\mathbf{k}) = \mathcal{E}'_0 + \frac{\hbar^2\mathbf{k}^2}{2M'} + \dots; \quad (7)$$

The energy of an unexcited crystal is taken as the zero level. Using the notation  $\omega_0 = \mathcal{E}_0/\hbar$  and keeping in mind that only frequencies  $\omega$  near  $\omega_0$  will be of importance, we can rewrite (6) as

$$\mu \approx \frac{2Mc^2}{\hbar\omega}\left(1 - \frac{\omega_0}{\omega}\right) \approx \frac{2Mc^2}{\hbar\omega_0}\frac{\omega - \omega_0}{\omega_0}, \quad \mu' \approx \frac{2M'c^2}{\hbar\omega'_0}\frac{\omega - \omega'_0}{\omega'_0} \quad (8)$$

Furthermore

$$b \equiv 8\pi Mc^2 a/\hbar^2\omega^3 \approx 8\pi Mc^2 a/\hbar^2\omega_0^3, \quad (9)$$

where  $a$  is a constant whose significance for the general case is given in Ref. 1. When we are concerned with the Frenkel type of exciton,<sup>2</sup> and when it is possible in zero approximation to write the wave function of the crystal as the product of the wave functions for the individual unit cells,  $a$  can be related to the oscillator strength  $f$  for the optical transition of a single cell by the equation

$$a = (e^2\hbar/2m)Nf, \quad (10)$$

where  $N$  is the number of cells in unit volume of the crystal and  $m$  is the mass of a free electron. From (9) and (10) by introducing the familiar constants  $a_0 = \hbar/me^2 = 0.529 \text{ \AA}$  and  $I = e^2/a_0 = 27.1 \text{ eV}$ , we finally obtain

$$b \approx \frac{4\pi Mc^2}{\mathcal{E}_0} \frac{(I/\mathcal{E}_0)^2}{(d/a_0)^3} f, \quad (11)$$

where  $d$  is the lattice constant.

In (4) " $\vartheta$ " is the contribution to the dielectric constant from virtual transitions to all excited states except the exciton state under consideration. If the other absorption bands are sufficiently distant from the band that is of interest here, " $\vartheta$ " can be regarded as independent of the frequency.

In solutions such as Eqs. (1) and (2), the magnetic field  $\mathbf{H}$  can be expressed in terms of the electric field  $\mathbf{E}$  by the equation  $\mathbf{H}_\pm = n[\mathbf{s} \times \mathbf{E}_\pm]$  of ordinary crystal optics. The electric induction is given by  $\mathbf{D}_\pm = n_\pm^2 \mathbf{E}_\pm$ . In solutions such as (3) we have  $\mathbf{H}_\parallel = \mathbf{D}_\parallel = 0$ .

The dispersion curves that correspond to (4) are represented by solid lines in Figs. 1 and 2, which depict the following numerical example:

$\mathcal{E}_0 = 2 \text{ eV}$ ,  $d/a_0 = 10$ ,  $\vartheta = 2$ ,  $f = 0.1$ . Fig. 1 corresponds to the case of  $M = m$ , for which we obtain  $b = 58400$ . Fig. 2 corresponds to the same  $M$  and  $b$  as in Fig. 1, but negative.

All previous attempts to construct a theory of light dispersion in crystals have tacitly assumed the inviolacy of Maxwell's phenomenological "ma-

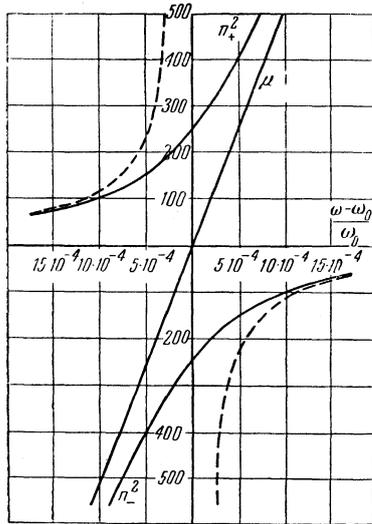


FIG. 1

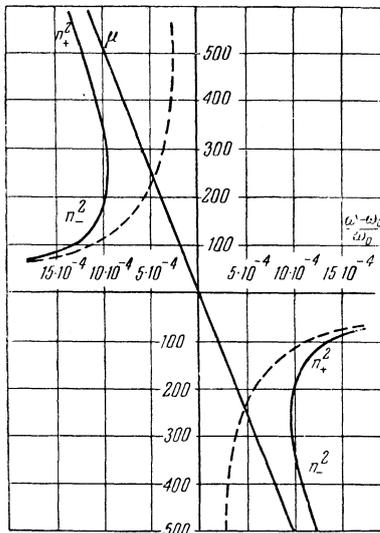


FIG. 2

terial" equations. We believe that this has led to incorrect results. Most of these attempts have resulted in a dispersion formula of the form (for electronic dispersion near an isolated narrow absorption band)

$$n^2 = \varepsilon - \frac{4\pi e^2 N f}{m(\omega^2 - \omega_0^2)} \approx \varepsilon - \frac{2\pi e^2 N f}{m\omega_0(\omega - \omega_0)} = \varepsilon - \frac{b}{\mu}. \quad (12)$$

For brevity we shall call this the ordinary dispersion formula, which is represented by a dashed line in Figs. 1 and 2. It will be shown subsequently that this formula is applicable only when light excitation is strongly associated with a lattice site (for large effective exciton mass), in which case the crystal can be regarded as a compressed gas.

As  $\omega$  moves away from  $\omega_0$  in either direction, we have the inequality  $4b/\mu^2 \ll 1$ , which in Eq. (4)

enables us to expand the radical in powers of  $b/\mu^2$ . The following asymptotic expressions result:

$$\begin{aligned} \text{for } \mu > 0 \quad n_+^2 &\approx \mu + b/\mu, \quad n_-^2 \approx \varepsilon - b/\mu, \\ \text{for } \mu < 0 \quad n_+^2 &\approx \mu + b/\mu, \quad n_-^2 \approx \varepsilon - b/\mu. \end{aligned} \quad (13)$$

It is thus seen that when the subscript of  $n_{\pm}^2$  agrees with the sign of  $\mu$ ,  $n_{\pm}^2$  approaches  $\mu$  asymptotically. When the subscript of  $n_{\pm}^2$  is the opposite of the sign of  $\mu$ ,  $n_{\pm}^2$  can be expressed asymptotically by Eq. (12). This can be seen in Figs. 1 and 2, where the solid curve approaches the dashed curve asymptotically. The difference between the new and the ordinary dispersion formula is more pronounced on steep portions of the dispersion curve than on shallow portions. Unlike ordinary dispersion curves the new curves have an inclined rather than a vertical straight asymptote. Therefore for all  $\omega$ ,  $n_+^2$ ,  $n_-^2$ , and  $n_{\parallel}^2$  remain finite, whereas by Eq. (12)  $n^2$  becomes infinite for  $\omega = \omega_0$ . In the ordinary formula a single value of  $n^2$  corresponds to each frequency; in the new formula there are three such values of  $n^2$ . If the atomic separation is continuously enlarged, thus increasing the lattice constant, we make the transition to a gas; this signifies an unlimited increase of the effective exciton mass  $M$ . Then, as is seen from Eq. (8), the inclined asymptote (the straight line  $\mu$ ) in the figures becomes vertical and the new dispersion curve goes over into the ordinary curve, as expected.

In Figs. 1 and 2 only real values of  $n^2$  are represented. When  $n^2 > 0$  the solution is an ordinary unattenuated monochromatic light wave. When  $n^2 < 0$ ,  $n$  is purely imaginary. This denotes that the electric and magnetic fields are gradually attenuated from the crystal surface inwards and cannot appreciably penetrate the crystal; in this case there is zero energy flow into the crystal because of the phase difference  $\pi/2$  between the electric and magnetic fields. Only for  $M < 0$ , since  $b < 0$  for  $\omega$  near  $\omega_0$ , the quantity under the radical in (4) becomes negative and  $n_{\pm}^2$  becomes complex (this is not shown in Fig. 2). In this region  $-b > (\mu - \varepsilon)^2/4$  and

$$\begin{aligned} n_{\pm} &= \left[ \frac{1}{4} (\mu + \varepsilon) + \frac{1}{2} \sqrt{-b + \mu\varepsilon} \right]^{1/2} \\ &\pm i \left[ \frac{-b - (\mu - \varepsilon)^2/4}{\mu + \varepsilon + 2\sqrt{-b + \mu\varepsilon}} \right]^{1/2}. \end{aligned} \quad (14)$$

We shall now show that when the ordinary dispersion formula (12) is applied to crystals there is sometimes considerable disagreement with experiment. According to (12) there is a region of

negative values of  $n^2$  beginning with  $\omega_0$  in the violet direction. This region extends to the frequency  $\omega_1$  which is determined from

$$\frac{\omega_1 - \omega_0}{\omega_0} = \frac{2\pi e^2 N f}{3 m \omega_0^2} = \frac{2\pi f}{3} \left( \frac{I}{\mathcal{E}_0} \right)^2 \left( \frac{a_0}{d} \right)^3. \quad (15)$$

Assuming  $f = 0.1$  and assuming the same values of the other parameters that were used in plotting the curves of Figs. 1 and 2, we obtain  $(\omega_1 - \omega_0)/\omega_0 = 0.058$ . When, for example,  $\omega_0 = 16200 \text{ cm}^{-1}$ , the width of the negative  $n^2$  region is given by  $\omega_1 - \omega_0 = 940 \text{ cm}^{-1}$  (assuming that there are no other strong bands on the violet side of the given absorption band). In this entire region the crystal would have to exhibit total reflection. This conflicts with experiment, since the widely used transillumination method of photographing crystal absorption spectra at low temperatures indicates that for the given oscillator strength the opaque region of a crystal plate is sometimes only 10 to 20  $\text{cm}^{-1}$ .

This difficulty does not arise in the proposed new theory of dispersion. Figure 1 shows that  $n_-^2$  is negative as far as the same frequency  $\omega_1$ , but that there is a second wave whose refractive index  $n_+$  is everywhere real. This wave passes freely through the crystal plate and generates an ordinary wave in the vacuum with exactly the same frequency as the original light. For a strictly quantitative determination of transparency under the new theory, Fresnel's equations must be extended to the case of the three waves appearing in a crystal.

## 2. FRESNEL EQUATIONS FOR THE VACUUM-TO-CRYSTAL BOUNDARY

Let the plane  $z = 0$  be the crystal surface; the crystal is located in the half-space  $z > 0$  while the half-space  $z < 0$  is a vacuum. Let the amplitudes of the electric fields of the waves, — the incident wave in the vacuum, the reflected wave in the vacuum and the three transmitted waves in the crystal, be denoted by  $\mathbf{A}$ ,  $\mathbf{R}$ ,  $\mathbf{E}_+$ ,  $\mathbf{E}_-$ , and  $\mathbf{E}_\parallel$ , respectively. The electric fields of the incident and reflected waves are given by

$$\begin{aligned} \mathbf{E} &= \mathbf{A} \exp \left\{ i\omega \left[ \frac{1}{c} (\mathbf{s} \cdot \mathbf{r}) - t \right] \right\}, \\ \mathbf{E} &= \mathbf{R} \exp \left\{ i\omega \left[ \frac{1}{c} (\tilde{\mathbf{s}} \cdot \mathbf{r}) - t \right] \right\}, \end{aligned} \quad (16)$$

and for the three transmitted waves they are given by Eqs. (1), (2), and (3). We denote the unit vectors of the normals to the wave fronts by  $\mathbf{s}$ ,  $\tilde{\mathbf{s}}$ ,  $\mathbf{s}_+$ ,  $\mathbf{s}_-$ , and  $\mathbf{s}_\parallel$ , respectively. We assume that  $y = 0$  is the plane of incidence. The incident wave forms the angle  $\varphi$  with the  $z$  axis, while the transmitted waves form the angles  $\psi_+$ ,  $\psi_-$ , and  $\psi_\parallel$ , as shown

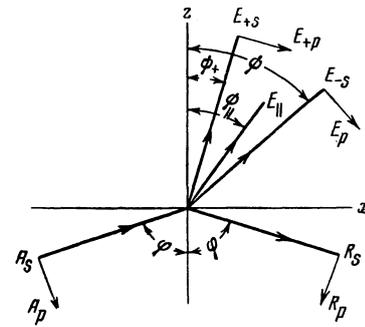


FIG. 3

in Fig. 3. Projections of the field amplitudes on the  $y$  axis will be denoted by the subscript  $s$ . The  $y$  axis in Fig. 3 is into the plane of the paper. Projections of the amplitudes on the plane of incidence will be denoted by the subscript  $p$ , with their positive direction indicated by the arrows in Fig. 3. The amplitude of the longitudinal wave will be considered positive in the direction of propagation of this wave.

As usual, the unit vectors of the normals and the amplitudes of all waves except the incident wave are determined from the continuity of the tangential projections of the electric and magnetic fields on the plane  $z = 0$ . For this purpose it is required, first of all, that in the plane  $z = 0$  there be agreement of the phases (the exponential indices) of all five waves for all values of  $x$ ,  $y$  and  $t$ . This leads to the relations

$$\begin{aligned} s_x = \tilde{s}_x = n_+ s_{+x} = n_- s_{-x} = n_\parallel s_{\parallel x}, \\ s_y = \tilde{s}_y = n_+ s_{+y} = n_- s_{-y} = n_\parallel s_{\parallel y} = 0, \end{aligned} \quad (17)$$

which express Snell's law and the requirement that all five waves possess the same frequency. The continuity of  $E_x$ ,  $H_y$ ,  $E_y$  and  $H_x$  is expressed by the following equations for the amplitudes:

$$\begin{aligned} (A_p - R_p) \cos \varphi \\ = E_{+p} \cos \psi_+ + E_{-p} \cos \psi_- + E_{\parallel} \sin \psi_\parallel, \end{aligned} \quad (18)$$

$$A_p + R_p = n_+ E_{+p} + n_- E_{-p}, \quad (19)$$

$$A_s + R_s = E_{+s} + E_{-s}, \quad (20)$$

$$(A_s - R_s) \cos \varphi = n_+ E_{+s} \cos \psi_+ + n_- E_{-s} \cos \psi_- \quad (21)$$

The continuity of the normal projection of the electric induction does not lead to new equations.

It is also necessary to satisfy the boundary condition derived in Ref. 1 for the specific dipole mo-

ment: in the plane  $z = 0$  we must have  $\mathbf{P}_1 = 0$ . In our notation this condition becomes

$$\frac{E_+}{n_+^2 - \mu} + \frac{E_-}{n_-^2 - \mu} - \frac{\partial}{b} E_{\parallel} = 0. \quad (22)$$

From the set of equations (18) to (21) and (22), the seven quantities  $R_s$ ,  $R_p$ ,  $E_{+s}$ ,  $E_{+p}$ ,  $E_{-s}$ ,  $E_{-p}$ , and  $E_{\parallel}$  are expressed in terms of  $A_s$  and  $A_p$  as follows:

s-components:

$$E_{\pm s} = u_{\pm} A_s, \quad R_s = (u_+ + u_- - 1) A_s, \quad (23)$$

$$u_{\pm} = 2 \left[ 1 + n_{\pm} \frac{\cos \psi_{\pm}}{\cos \varphi} - q^{\pm 1} \left( 1 + n_{\mp} \frac{\cos \psi_{\mp}}{\cos \varphi} \right) \right]^{-1}, \quad (24)$$

$$u_- = -q u_+, \quad q \equiv \frac{n_-^2 - \mu}{n_+^2 - \mu} = \frac{\partial - n_+^2}{\partial - n_-^2},$$

$$n_{\pm} = \frac{\sin \varphi}{\sin \psi_{\pm}}, \quad n_{\parallel} = \sqrt{\mu} = \frac{\sin \varphi}{\sin \psi_{\parallel}}; \quad (25)$$

p-components:

$$E_{\pm p} = v_{\pm} A_p, \quad R_p = (v_+ n_+ + v_- n_- - 1) A_p, \quad (26)$$

$$E_{\parallel} = \frac{\sin(\psi_- - \psi_+)}{\cos(\psi_- - \psi_{\parallel})} \frac{b v_+}{\partial (n_+^2 - \mu)} A_p, \quad (27)$$

$$v_{\pm} = 2 \left[ \frac{\cos \psi_{\pm}}{\cos \varphi} + n_{\pm} + \frac{b \sin \psi_{\parallel} \sin(\psi_{\mp} - \psi_{\pm})}{\partial (n_{\pm}^2 - \mu) \cos \varphi \cos(\psi_{\mp} - \psi_{\parallel})} - q^{\pm 1} \frac{\cos(\psi_{\pm} - \psi_{\parallel})}{\cos(\psi_{\mp} - \psi_{\parallel})} \left( \frac{\cos \psi_{\mp}}{\cos \varphi} + n_{\mp} \right) \right]^{-1}, \quad (28)$$

where

$$v_- = -q \frac{\cos(\psi_+ - \psi_{\parallel})}{\cos(\psi_- - \psi_{\parallel})} v_+. \quad (29)$$

It can be shown that when  $M > 0$ , for example, as  $\omega$  changes toward the red from  $\omega_0$ , the (+) wave acquires all the properties of the ordinary wave, while the amplitude of the anomalous (-) wave approaches zero ( $|q|$  becomes  $\ll 1$ ). The formulas given above then become the ordinary Fresnel equations with  $n_+$  as the refractive index. Similarly, as  $\omega$  changes toward the violet from  $\omega_0$ , the (-) wave becomes the ordinary wave while the amplitude of the anomalous (+) wave approaches zero ( $|q|$  becomes  $\gg 1$ ). The above formulas then become the ordinary Fresnel equations with  $n_-$  as the refractive index, and the (-) wave becomes the transmitted wave.

The coefficient of reflection from the crystal surface into the vacuum is

$$\text{for the s-component } r_s = |R_s|^2 / |A_s|^2 = |u_+ + u_- - 1|^2, \quad (30)$$

$$\text{for the p-component } r_p = |R_p|^2 / |A_p|^2$$

$$= |v_+ n_+ + v_- n_- - 1|^2.$$

In the special case of normal incidence

$$r_s = r_p = r = \left| \frac{n_+ - 1 - q(n_- - 1)}{n_+ + 1 - q(n_- + 1)} \right|^2. \quad (31)$$

We shall now consider the passage of light through the boundary between the vacuum and the crystal with the incident wave striking the crystal surface from within. The longitudinal, (+), and (-) waves will be considered separately. As previously, Snell's law applies to all cases; the product of the refractive index and the sine of the angle formed with the  $z$  axis is identical for all waves.

#### a. For an Incident Longitudinal Wave.

We retain all of the notation of Fig. 3 with the following necessary changes. In the half-space  $z < 0$  there now exists only one wave propagated away from the surface with amplitude  $R$  ( $A = 0$ ). In the half-space  $z > 0$  the negative- $x$  quadrant contains an additional longitudinal wave whose direction forms the angle  $\psi_{\parallel}$  with the  $z$  axis. The amplitude of the latter wave will be denoted by  $A_{\parallel}$ . The boundary conditions for the s components of the amplitudes are derived directly from Eqs. (20) and (21) by inserting  $A_s = 0$ . We require in addition the s projection of the vector equation (22). The solution of these equations is obtained from (23) by inserting  $A_s = 0$ . Thus  $E_{+s} = E_{-s} = R_s = 0$ .

For the p-components of the amplitudes we obtain the following equations which express the continuity of  $E_x$  and  $H_y$ :

$$(A_{\parallel} + E_{\parallel}) \sin \psi_{\parallel} + E_{+p} \cos \psi_+ + E_{-p} \cos \psi_- = -R_p \cos \varphi, \quad (32)$$

$$n_+ E_{+p} + n_- E_{-p} = R_p. \quad (33)$$

In addition, the condition  $\mathbf{P}_1 = 0$  on the crystal surface<sup>1</sup> leads to

$$\frac{E_{+p} \cos \psi_+}{n_+^2 - \mu} + \frac{E_{-p} \cos \psi_-}{n_-^2 - \mu} - \frac{\partial}{b} (E_{\parallel} + A_{\parallel}) \sin \psi_{\parallel} = 0, \quad (34)$$

$$\frac{E_{+p} \sin \psi_+}{n_+^2 - \mu} + \frac{E_{-p} \sin \psi_-}{n_-^2 - \mu} + \frac{\partial}{b} (E_{\parallel} - A_{\parallel}) \cos \psi_{\parallel} = 0. \quad (35)$$

The solution of Eqs. (32) to (35) is

$$E_{\pm p} = \omega_{\pm} A_{\parallel}, E_{\parallel} = \omega_{\parallel} A_{\parallel}, R_p = (n_+ \omega_+ + n_- \omega_-) A_{\parallel}, \quad (36)$$

where

$$\omega_{\pm} = - \frac{\sin 2\psi_{\parallel} \left[ \frac{\vartheta}{b} \left( \frac{\cos \psi_{\mp}}{\cos \varphi} + n_{\mp} \right) (n_{\mp}^2 - \mu) + \frac{\cos \psi_{\mp}}{\cos \varphi} \right]}{\left( \frac{\cos \psi_{\pm}}{\cos \varphi} + n_{\pm} \right) \cos(\psi_{\parallel} - \psi_{\mp}) - q^{\pm 1} \left( \frac{\cos \psi_{\mp}}{\cos \varphi} + n_{\mp} \right) \cos(\psi_{\parallel} - \psi_{\pm}) + \frac{b}{\vartheta} \frac{\sin \psi_{\parallel} \sin(\psi_{\mp} - \psi_{\pm})}{\cos \varphi (n_{\pm}^2 - \mu)}}, \quad (37)$$

$$\omega_{\parallel} = \frac{\left( \frac{\cos \psi_+}{\cos \varphi} + n_+ \right) (n_+^2 - \mu) \cos(\psi_{\parallel} + \psi_-) - \left( \frac{\cos \psi_-}{\cos \varphi} + n_- \right) (n_-^2 - \mu) \cos(\psi_{\parallel} + \psi_+) - \frac{b}{\vartheta} \frac{\sin \psi_{\parallel} \sin(\psi_- - \psi_+)}{\cos \varphi}}{\left( \frac{\cos \psi_{\pm}}{\cos \varphi} + n_{\pm} \right) (n_{\pm}^2 - \mu) \cos(\psi_{\parallel} - \psi_{\pm}) - \left( \frac{\cos \psi_{\mp}}{\cos \varphi} + n_{\mp} \right) (n_{\mp}^2 - \mu) \cos(\psi_{\parallel} - \psi_{\mp}) + \frac{b}{\vartheta} \frac{\sin \psi_{\parallel} \sin(\psi_- - \psi_+)}{\cos \varphi}} \quad (38)$$

As explained in Ref. 1, the longitudinal wave is the electric field of an exciton wave excited in the crystal. Equations (36) to (38) show that the longitudinal (exciton) wave is only partly reflected from the crystal surface ( $|w_{\parallel}|^2 \leq 1$ ); the rest of its energy is expended for the generation of two electromagnetic waves directed into the crystal [the (+) and (-) waves] and of an additional wave directed into the vacuum. In other words, excitons which reach the crystal surface emit light.

For normal incidence  $w_{\parallel} = 1$  and  $w_{\pm} = 0$ .

#### b. For an Incident (+) Wave.

This case differs from the preceding one only in that the longitudinal wave is replaced by an incident (+) wave of amplitude  $A_+$ . The directions of all waves are shown in Fig. 4. The continuity

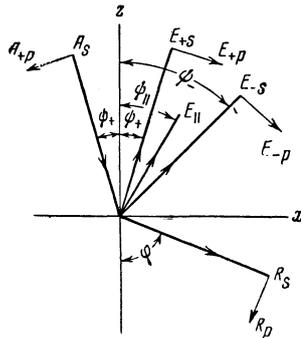


FIG. 4

conditions for  $E_x$ ,  $H_y$ ,  $E_y$ , and  $H_x$  become

$$-R_p \cos \varphi = (E_{+p} - A_{+p}) \cos \psi_+ + E_{-p} \cos \psi_- + E_{\parallel} \sin \psi_{\parallel}, \quad (39)$$

$$R_p = n_+ (E_{+p} + A_{+p}) + n_- E_{-p}, \quad (40)$$

$$R_s = A_{+s} + E_{+s} + E_{-s}, \quad (41)$$

$$R_s \cos \varphi = n_+ (A_{+s} - E_{+s}) \cos \psi_+ - n_- E_{-s} \cos \psi_-. \quad (42)$$

The condition  $\mathbf{P}_{\parallel} = 0$  in the  $z = 0$  plane<sup>1</sup> leads to

$$\frac{E_+ + A_+}{n_+^2 - \mu} + \frac{E_-}{n_-^2 - \mu} - \frac{\vartheta}{b} E_{\parallel} = 0. \quad (43)$$

The solution of Eqs. (39) to (43) is for s-components:

$$E_{+s} = \frac{n_+ \cos \psi_+ + q n_- \cos \psi_- - (1 - q) \cos \varphi}{n_+ \cos \psi_+ - q n_- \cos \psi_- + (1 - q) \cos \varphi} A_{+s}, \quad (44)$$

$$E_{-s} = - \frac{2q n_+ \cos \psi_+}{n_+ \cos \psi_+ - q n_- \cos \psi_- + (1 - q) \cos \varphi} A_{+s}, \quad (45)$$

$$R_s = \frac{2(1 - q) n_+ \cos \psi_+}{n_+ \cos \psi_+ - q n_- \cos \psi_- + (1 - q) \cos \varphi} A_{+s}; \quad (46)$$

for p-components:

$$E_{\parallel} = - \frac{bU}{\vartheta (n_+^2 - \mu)} A_{+p},$$

$$U = \frac{\sin 2\psi_+ \left[ (1 - q) \frac{\cos \psi_-}{\cos \varphi} - q n_- \right]}{\left( \frac{\cos \psi_+}{\cos \varphi} + n_+ \right) \cos(\psi_{\parallel} - \psi_-) - q \left( \frac{\cos \psi_-}{\cos \varphi} + n_- \right) \cos(\psi_{\parallel} - \psi_+)} \rightarrow \frac{+2n_+ \sin \psi_- \cos \psi_+}{b \sin \psi_{\parallel} \sin(\psi_- - \psi_+)} + \frac{1}{\vartheta (n_+^2 - \mu) \cos \varphi}, \quad (47)$$

$$E_{+p} = \frac{\sin(\psi_- + \psi_+) - U \cos(\psi_{\parallel} - \psi_-)}{\sin(\psi_- - \psi_+)} A_{+p}, \quad (48)$$

$$E_{-p} = q \frac{U \cos(\psi_{\parallel} - \psi_+) - \sin 2\psi_+}{\sin(\psi_- - \psi_+)} A_{+p}, \quad (49)$$

$$R_p = \{2n_+ \sin \psi_- \cos \psi_+ - q n_- \sin 2\psi_+ - U [n_+ \cos(\psi_{\parallel} - \psi_-) - q n_- \cos(\psi_{\parallel} - \psi_+)]\} A_{+p} / \sin(\psi_- - \psi_+). \quad (50)$$

**c. For an Incident (-) Wave.**

This is entirely analogous to case b with the single difference that the (+) and (-) waves have exchanged roles. Therefore all equations and solutions for this case are obtainable from Eqs. 39 to 50 simply by interchanging the subscripts + and - and replacing  $q$  with  $1/q$ .

**3. TRANSMISSION OF LIGHT THROUGH A PLANE-PARALLEL PLATE**

We take the planes  $z = 0$  and  $z = l$  as the surfaces of the plate. We limit ourselves to a consideration of normal incidence, where the incident and all secondary waves propagate parallel to the  $z$  axis. Then Eqs. 27 and 47 show that longitudinal waves do not arise in the crystal, but that there are four transverse waves, whose electric fields will be denoted as follows:

$$\begin{aligned} E_+ e^{i(k_+ z - \omega t)}, \quad E_- e^{i(k_- z - \omega t)}, \\ E'_+ e^{i(-k_+ z - \omega t)}, \quad E'_- e^{i(-k_- z - \omega t)}, \end{aligned} \quad (51)$$

where

$$k_{\pm} = n_{\pm} \omega / c.$$

The electric fields of the incident wave, the wave reflected into the region  $z < 0$ , and the wave transmitted into the region  $z > l$  will be denoted respectively by

$$A e^{i(k_+ z - \omega t)}, \quad R e^{i(-k_+ z - \omega t)}, \quad D e^{i(k_+ z - \omega t)} \quad (k_0 = \omega / c). \quad (52)$$

Continuity of the tangential electric and magnetic components on the surface  $z = 0$  furnishes the equations

$$A + R = E_+ + E_- + E'_+ + E'_-, \quad (53)$$

$$A - R = n_+ (E_+ - E'_+) + n_- (E_- - E'_-). \quad (54)$$

The analogous condition on the surface  $z = l$  gives

$$E_+ e^{i k_+ l} + E_- e^{i k_- l} + E'_+ e^{-i k_+ l} + E'_- e^{-i k_- l} = D e^{i k_+ l}, \quad (55)$$

$$\begin{aligned} n_+ (E_+ e^{i k_+ l} - E'_+ e^{-i k_+ l}) \\ + n_- (E_- e^{i k_- l} - E'_- e^{-i k_- l}) = D e^{i k_+ l} \end{aligned} \quad (56)$$

The condition  $\mathbf{P}_1 = 0$  on the surfaces  $z = 0$  and  $z = l$  gives, respectively,

$$E_- + E'_- = -q (E_+ + E'_+), \quad (57)$$

$$E_- e^{i k_- l} + E'_- e^{-i k_- l} = -q (E_+ e^{i k_+ l} + E'_+ e^{-i k_+ l}). \quad (58)$$

Equations 53 to 58 can be used to express the secondary wave amplitudes  $E_+$ ,  $E_-$ ,  $E'_+$ ,  $E'_-$ ,  $R$ , and  $D$  in terms of the incident wave amplitude  $A$ . From (53), (55), (57), and (58) we obtain

$$\begin{aligned} E_+ &= \frac{i}{2} \frac{(A + R) e^{-i k_+ l} - D e^{i k_+ l}}{(1 - q) \sin k_+ l}, \\ E_- &= \frac{i}{2} \frac{(A + R) e^{-i k_- l} - D e^{i k_- l}}{(1 - 1/q) \sin k_- l}, \\ E'_+ &= -\frac{i}{2} \frac{(A + R) e^{i k_+ l} - D e^{i k_+ l}}{(1 - q) \sin k_+ l}, \\ E'_- &= -\frac{i}{2} \frac{(A + R) e^{i k_- l} - D e^{i k_- l}}{(1 - 1/q) \sin k_- l}. \end{aligned} \quad (59)$$

Substitution of (59) into (54) and (56) gives

$$D = \frac{2iG e^{-i k_+ l}}{(1 + iF)^2 + G^2} A; \quad R = \frac{1 + F^2 - G^2}{(1 + iF)^2 + G^2} A, \quad (60)$$

where

$$F = \frac{n_+}{1 - q} \cot k_+ l + \frac{n_-}{1 - 1/q} \cot k_- l;$$

$$G = n_+ / (1 - q) \sin k_+ l + n_- / (1 - 1/q) \sin k_- l. \quad (61)$$

Thus far it has been assumed that  $\mu$  and  $\epsilon$  are real. Then  $n_+$ ,  $n_-$ , and  $n_{\parallel}$  for  $M < 0$  are either purely real or purely imaginary. This denotes complete absence of light absorption in the plate. For  $M < 0$ , in the frequency region to which Eq. 14 is applicable,  $n_+$  and  $n_-$  are complex conjugates. It can be shown that in this case no light will be absorbed in the plate. It must be emphasized that in virtue of the assumption that the lifetime of the exciton state is determined only by interaction with the electromagnetic field (emission), there can be no exciton absorption of light for any magnitude of the unit-cell oscillator strength  $f$ . This becomes obvious from consideration of the stationary state in which an electromagnetic wave entering the crystal for an infinitely long period does not raise the average quantum-mechanical energy level of the crystal. Therefore all of the admitted light energy must be re-emitted by the crystal plate. Unlike a gas, however, the crystal will emit radiation only parallel to the  $z$  axis. Radiation in the positive  $z$  direction is called a transmitted wave, while radiation in the negative  $z$  direction is called a reflected wave. The sum of the transmitted and reflected intensities equals the incident intensity.

Exciton absorption of light in a crystal occurs when the system makes transitions from exciton states produced by light to any state other than the original state. When these transitions are

accompanied by emission, Roman scattering of the primary light occurs. When these transitions are performed thermally, with the excitation of thermal vibrations, ordinary light absorption occurs. It is thus clear that a theoretical study of light absorption must take into account the finite exciton lifetime which results from all possible transitions except a transition to the original state of the system. The finite exciton lifetime can be taken into account formally by a small imaginary correction to the exciton energy  $\mathcal{E}(k)$  in Ref. 1. A corresponding imaginary correction appears in  $\mu$ . All of the equations derived above remain in effect and will describe the case of an absorbing crystal since  $n_+$ ,  $n_{\pm}$ , and  $n_{\parallel}$  now have new complex values.

A special article will be devoted to exciton absorption of light. We shall here confine ourselves to negligibly small absorption, which occurs when  $n_+$  and  $n_-$  are real or pure imaginary quantities (not necessarily both simultaneously) and also when  $n_- = n_-^*$ . In such cases Eq. 61 shows that  $F$  and  $G$  are real. The transmission coefficient  $\delta$  and the reflection coefficient  $\rho$  are

$$\delta = \frac{|D|^2}{|A|^2} = \frac{4G^2}{(1+F^2-G^2)^2+4G^2},$$

$$\rho = \frac{|R|^2}{|A|^2} = \frac{(1+F^2-G^2)^2}{(1+F^2-G^2)^2+4G^2}. \quad (62)$$

Hence we obtain  $\delta + \rho = 1$ , which proves the absence of light absorption.

Interference transmission maxima, when  $\delta = 1$  and  $\rho = 0$ , occur at frequencies determined by the condition  $1 + F^2 - G^2 = 0$ . Transmission minima, when  $\delta = 0$  and  $\rho = 1$ , occur at frequencies determined by  $G = 0$ .

Equations 57 and 58 show that for  $q \rightarrow 0$  both  $(-)$  waves disappear in the crystal, and that for  $|q| \rightarrow \infty$  both  $(+)$  waves disappear. In both instances Eqs. 53 to 58 are the same as in ordinary crystal optics. Therefore when in Eqs. 59 to 62 we make the formal transition to the limit  $q \rightarrow 0$  or  $|q| \rightarrow \infty$ , we obtain the equations of ordinary crystal optics. Equations 60 and 62 then remain unchanged but Eq. 61 is replaced by

$$F_0 = n \cot kl, \quad G_0 = n / \sin kl, \quad k = \omega n / c. \quad (63)$$

We now turn to the question of crystal transparency in the frequency region  $\omega_0 - \omega_1$ , where according to the ordinary dispersion equation (12)  $n$  is pure imaginary. From (63) we now obtain  $G_0 = |n| / \sinh |k| l$ . Consequently,  $G_0$  decreases exponentially with  $l$ , and when the thickness of the plate is considerably greater than the wavelength

we have practically  $G_0 \approx 0$  and  $\delta \approx 0$ . This represents total reflection in the entire interval from  $\omega_0$  to  $\omega_1$ , which, as noted in Sec. 1, conflicts with experiment. The aforementioned difficulty of ordinary crystal optics does not arise in the proposed theory, where  $G$  is expressed by (61). Here we obtain  $G \approx 0$  only when  $M < 0$  in the frequency region where both  $n_+$  and  $n_-$  are pure imaginary (Fig. 2). When  $M > 0$ , owing to the presence of second light with a real refractive index  $n_+$ , the first term in the expression for  $G$  is large. Therefore  $G \neq 0$  and  $\delta \neq 0$ . Thus the plate can be transparent in the frequency interval  $\omega_0 - \omega_1$ .

#### 4. POSSIBLE EXPERIMENTAL TESTS OF THE PROPOSED THEORY

In selecting crystals and absorption bands for testing of the theory it must be borne in mind that these must be exciton absorption bands with the term "exciton" understood according to the definition at the beginning of Ref. 1. Extremely low temperatures are preferable because the theory has been developed, strictly speaking, for absolute zero. Although the theory can be applied to very low temperatures above zero, suitable criteria have not been established. The absorption bands must not be adjacent to other strong bands. It is desirable to begin with cubic crystals, in which the effects under investigation are not complicated by birefringence.

It is desirable to investigate the case of small effective exciton mass, where the straight asymptotes in Figs. 1 and 2 show greater inclination away from the vertical. In such cases second and third light possess appreciable amplitudes in a broader frequency range. Narrow absorption bands must be selected, which are usually associated with photo-transitions that do not induce multiple phonon production. Narrow bands are also more suitable because they permit more complete determination of the dispersion curves.

In using previously published experimental results we can trust the estimate of the elementary cell oscillator strength  $f$  based on the low-frequency tail of the dispersion curve, since the ordinary dispersion equation (12) used for this purpose approaches Eq. 4 asymptotically. But the oscillator strength  $f$  cannot be evaluated from the "absorption curve area" by the ordinary theory, since the proposed theory includes an entirely different equation.

We shall now give a far from complete list of the experiments that could be performed to test the proposed theory.

1. Determination of the cases of crystal transparency in a considerable portion of the frequency interval from  $\omega_0$  to  $\omega_1$  [see Eq. (15)].

2. Plotting of dispersion curves such as those in Figs. 1 and 2, by means of experimental determination of the refractive indices of all three waves. Some possible methods are:

a. An interferometer is used to divide the original light beam into two beams; one of the beams passes through a plate of the investigated crystal, after which it interferes with the other beam. Study of this interference enables us to determine the change of amplitude and phase of the light wave as a result of passage through the plate, i.e., the complex quantity  $D/A$  (in the notation of Sec. 3). In the ordinary theory this determines the single refractive index but in the proposed theory the fraction in the first equation of (60) is determined, thus giving an equation with the three unknowns  $n_+$ ,  $n_-$ ,  $\mu$ . By performing the measurement for three thicknesses of the plate, we obtain three equations to determine the three unknown quantities.

It is also possible to make an interference study of the light reflected from the crystal plate, thus determining the fraction in the second equation of (60) and giving an equation for the determination of  $n_+$ ,  $n_-$ , and  $\mu$ . Thus two relations can be obtained with a plate of any given thickness.

b. The refractive indices can be determined by sending light through a prism made of the given crystal. In Sec. 2 it was shown that a beam entering the prism is split into three beams (see Fig. 3). The three beams emerge from the crystal prism at different angles with the primary beam which can be measured in the usual way to give all three refractive indices by means of Snell's law. Detection of the three beams emerging from a prism would be a direct proof of the existence of second and third light in a cubic crystal.

Since the frequency of the light is in or near the absorption region, the prism must be a very thin and sharp wedge to insure sufficient transparency. It must be remembered that the refractive indices of second and third light can be very large, amounting to some tens or even hundreds. Therefore if these beams are to emerge from the prism instead of undergoing total internal reflection, their direction within the prism must form a very small angle with the normal to the plane of emergence. After emergence, the beams should be sought at a large angle to the normal.

When the primary beam is normal to the entrance plane, no longitudinal wave arises in the prism. When the direction of a longitudinal wave

propagating in the prism is normal to the exit plane of the prism, it undergoes total internal reflection and thus excites no electromagnetic wave in the vacuum. If the refractive index of any one of the three waves is imaginary, that wave will pass through the prism only if the thickness of the latter does not exceed the wavelength in order of magnitude. It is especially interesting to observe the frequency region in which  $|q|$  is of the order of unity, in which case the amplitude of second light is of the same order as that of ordinary light.

As a control over any method of determining the refractive indices, the following relation, which is derived from (4), should be verified:

$$n_+^2 + n_-^2 - \mu = \epsilon, \quad (64)$$

where  $\epsilon$  should not depend on the frequency if there are no close-lying absorption bands.

3. Interference investigation of a beam reflected from a semi-infinite crystal makes it possible to determine the changes of phase and amplitude upon reflection, i.e., the complex quantities  $R_s/A_s$  and  $R_p/R_p$ . By equating these quantities and their theoretical values as given by Eqs. (23) and (26), we obtain in each instance an equation relating  $n_+$ ,  $n_-$ , and  $\mu$ . When the measurement is performed three times at different angles of incidence, or with different polarizations, we obtain three equations for  $n_+$ ,  $n_-$ , and  $\mu$ , from which the refractive indices can be determined. When the coefficient  $b$  is unknown, it is best to confine the investigation to  $s$ -polarization, since Eqs. (23) and (24) do not contain  $b$ .

4. When any one of the aforementioned methods is used to determine  $n_+$ ,  $n_-$ , and  $\mu$ , the value of  $\mu$  can be used in (8) to calculate the effective exciton mass  $M$ . Then  $b$  can be calculated from (4), using, for example, the value of  $n_+^2$ . Substitution of the result into (4) for  $n_-^2$  serves as a check.

5. By analogy with Brewster's law in the ordinary theory, the proposed theory leads to an angle of incidence at which the  $p$ -component is completely unreflected, so that the reflected beam is strictly polarized in the  $s$ -direction. Equation (26) shows that this occurs when  $v_+n_+ + v_-n_- - 1 = 0$ . This equation can also be used to verify the theory after experimental determination of the refractive indices and angle of incidence at which total polarization of the reflected wave is observed.

Exciton absorption of light will be considered in another article.

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## THE MOTIONS OF ROTATING MASSES IN THE GENERAL THEORY OF RELATIVITY

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The relativistic equations of translational and rotational motion for spherically symmetrical rotating bodies, developed in a previous paper,<sup>1</sup> have been integrated. Some novel relativistic effects, due to the proper rotations of the bodies, appear and are discussed.

### 1. INTRODUCTION

IN an article by one of the authors<sup>1</sup> the equations of translational and rotational motion for spherically symmetrical rotating bodies were derived from Einstein's gravitational equations. In the present paper we shall study the solutions of these equations. In view of the well-known difficulties in the general problem of celestial mechanics, we shall limit ourselves here to a study of the two-body problem. In their well-known paper,<sup>2</sup> Thirring and Lense studied the relativistic effects of rotation as applied to the very simple (though important) case of a very light, non-rotating body moving in the field of a massive rotating body, by making use of the properties of geodesics. In our problem, however, both bodies are treated on an equal footing — they may be of comparable mass, and each may rotate about its own axis.

Let  $a^i$  and  $b^i$ ,  $m_a$  and  $m_b$ ,  $M_a^{ik}$  and  $M_b^{ik}$  be the coordinates, masses, and proper rotational moments of the two bodies, and let  $r$  be the distance between them and  $\gamma$  the Newtonian gravitational constant. In Ref. 1 the equations

$$\ddot{a}^i - \left( \frac{\gamma m_b}{r} \right)_{,a^i} = F_a^i + D_a^i \quad (1.1)$$

were derived for the translational motion, and

$$M_a^{ih} = L_a^{ih} \quad (1.2)$$

for the proper rotation. (Analogous equations hold for body b). Here  $F_a^i$  is the relativistic correction to the Newtonian force when the rotation of the body is neglected; it has been discussed by numerous authors.<sup>3,4,5</sup>  $D_a^i$  is the relativistic correction due to the rotation derived in Ref. 1 (cf. Eq. (5.6) of that paper).  $L_a^{ik}$  is an abbreviation for the right-hand side of equation (6.2) of Ref. 1. We shall not repeat here the complicated expressions for  $F_a^i$ ,  $D_a^i$ , and  $L_a^{ik}$ .

In the Newtonian approximation,  $F_a^i = D_a^i = L_a^{ik} = 0$ , and we obtain the familiar solution, with  $r \equiv |\mathbf{a} - \mathbf{b}|$ ,

$$\frac{1}{r} = \frac{1}{p} (1 + e \cos \varphi), \quad (1.3)$$

$$M_a^{ih} = \text{const}, \quad M_b^{ih} = \text{const}, \quad (1.4)$$

subject to the conservation laws

$$M_1 = M_2 = 0, \quad M_3 \equiv r^2 \dot{\varphi} = (\gamma m p)^{1/2} = \text{const}, \quad (1.5)$$

$$E \equiv \frac{1}{2} v^2 - \frac{\gamma m}{r} = -\frac{\gamma m}{2a} = \text{const}. \quad (1.6)$$

Here  $e$  is the eccentricity of the orbit, if we take  $e < 1$ ;  $p$  is a parameter,  $a$  is the major semi-axis of the relative orbit,  $v$  is the relative velocity, and  $m \equiv m_a + m_b$ .