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**THEORY OF BANDS
AND DYNAMICAL SCATTERING
OF PARTICLES BY LARGE CRYSTALS**

by

G. BLAESSER

1972



Joint Nuclear Research Centre
Ispra Establishment - Italy
Physics Division

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ABSTRACT

Well-known expressions of the dynamical scattering theory are derived by the analogy with the mathematical similar situation of the propagation of electrons in a periodic potential. It is our hope that this similarity between the theory of electron bands and the theory of dynamical scattering can be used for the discussion of more complicated problems (for example: the dynamical scattering in the presence of impurities).

KEYWORDS

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CRYSTAL LATTICES
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1. Introduction

It is well known that usual kinematic scattering theory can be applied only to the case of small thickness - where multiple scattering events can be neglected - while in the case of large samples one has to use the so-called dynamical scattering theory (1,2,3,4).

In the standard texts dynamical scattering theory is derived for the particular case of X-ray scattering while taking into account all the polarization properties of electromagnetic waves by using the mathematical apparatus of classical Maxwellian theory. This derivation tends to obscure the essential physical features by the additional complications arising from these polarization effects.

It is therefore interesting to derive the essential properties of dynamical scattering for the case of scalar (i.e. spinless) particles since the polarization effects are not essential to the main results of dynamical scattering theory.

In the present paper the results of dynamical scattering theory are rederived along these lines; by this procedure it is also seen that dynamical scattering theory bears a striking resemblance to the theory of electron bands in crystals : it is therefore hoped that this similarity can be used to extend some results of the theory of electron bands to problems in dynamical scattering . This could be especially important in case where we have to study dynamical scattering in the presence of impurities, phonon interactions and nonlinearities.

2. The Propagation of Particles in an Infinite Ideal Crystal

Since the potential $V(\underline{r})$ which describes the interaction between the propagating particles of mass μ and the crystal atoms has to the symmetry of the crystal the Hamilton operator $H = T + V$ ($T = (\hbar^2/2\mu)\nabla^2 =$ kinetic energy operator) of these particles is invariant with respect to the space group \mathcal{G} of the crystal and in particular with respect to its subgroup \mathcal{T} of lattice translations \underline{t} , where the operators \underline{t} are defined by

$$1- \quad \underline{t}_{\underline{\alpha}} \varphi(\underline{r}) = \varphi(\underline{r} + \underline{\alpha})$$

and the $\underline{\alpha}$ are all vectors of the form

$$2- \quad \underline{\alpha} = \sum_{i=1}^3 m_i \underline{a}_i$$

where m_1, m_2 and m_3 are arbitrary integers, and $\underline{a}_1, \underline{a}_2$ and \underline{a}_3 are the Bravais vectors of the lattice (which define the elementary cell). Thus the invariance of $V(\underline{r})$ with respect to \mathcal{T} means that

$$3- \quad \underline{t}_{\underline{\alpha}} V(\underline{r}) = V(\underline{r} + \underline{\alpha}) = V(\underline{r})$$

Since H and the operators $\underline{t}_{\underline{\alpha}}$ commute, the eigenstates of H (i.e. the solutions of the Schrödinger equation $H\psi = E\psi$) must be representations of \mathcal{T} . As it is well known (5) all representations of \mathcal{T} can be written in the form of Bloch functions

$$4- \quad \psi(\underline{r}) = u_{\underline{k}}(\underline{r}) \exp(i\underline{k} \cdot \underline{r})$$

with periodic functions $u_{\underline{k}}(\underline{r})$:

$$5- \quad \underline{t}_{\underline{\alpha}} u_{\underline{k}}(\underline{r}) = u_{\underline{k}}(\underline{r} + \underline{\alpha}) = u_{\underline{k}}(\underline{r})$$

The vector \underline{k} is the parameter which distinguishes the different representations $\Gamma_{\underline{k}}$; within the first Brillouin zone different values of \underline{k} correspond to inequivalent representations.

These facts are well known for the case of electrons in a lattice. The dependence of the energy eigenvalues on

the representations is expressed in the dispersion relation $E = E(\underline{k})$ and exhibits the band structure. Here, however, we nowhere assumed that the propagating particles are electrons. Therefore these results are perfectly general and apply to all particles propagating in an ideal crystal.

In the usual treatment of the theory of electron bands (5) a first orientation about the band structure is obtained by the quasi-free electron approximation: One starts with the free-electron approximation, which neglects all interactions between the electrons and the lattice. The wave functions are the plane waves

$$6- \psi_{1,\underline{k}}(\underline{r}) = \exp \left[i (\underline{k} - \underline{K}_1) \underline{r} \right]$$

and the corresponding dispersion relation is

$$7- E_1(\underline{k}) = (\hbar^2/2\mu) (\underline{k} - \underline{K}_1)^2$$

The index 1 enumerates all vectors \underline{K}_1 of the reciprocal lattice; therefore $\exp(i\underline{K}_1 \cdot \underline{\alpha}) = 1$, so that the plane waves (6) are really Bloch functions with a periodic part $\exp(-i\underline{K}_1 \cdot \underline{r})$. The quasi-free electron approximation is now obtained from the free electron approximation by considering the interaction only at the points where we have accidental degeneracies in the free electron model. In this way the dispersion curves of the free electron case (fig. 1a) go over into the corresponding curves for the quasi-free electron case (fig. 1b). If we draw the contours of constant energy in two dimensions and apply the same perturbation procedure we obtain the effect shown in Fig. 2 for the particular case of the interaction of the two waves characterized by $\underline{K}_0 = (000)$ and $\underline{K}_1 = (2\pi/a)(100)$. Considering the influence of the other points \underline{K}_1 of the reciprocal lattice, the constant energy curves in two dimensions have a form like the one shown in Fig. 3. These curves are the (k_x, k_y) - sections of the three-dimensional constant energy surfaces; plots like fig. 3 are well known in the theory of the Fermi surface (which is nothing else than the constant energy surface for which the energy equals the Fermi level).

In the dynamical theory of scattering we can also use the quasi-free particle approximation. The surfaces $E = \text{const}$ will be called "dispersion surfaces" since they give the dispersion relation $E = E_1(\underline{k})$ in graphical form. It should be noted that for $\sqrt{2\mu E} > 2\pi\hbar/a$ the dispersion surface surrounds \underline{K}_0 and \underline{K}_1 as in Fig.4 In such a case also higher order waves could be important for its construction.

The separation of the energy levels which is illustrated in Fig.1 can easily be calculated from the Schrödinger equation in K-space. The periodic functions $u_{\underline{k}}(\underline{r})$ and $V(\underline{r})$ can be written as Fourier series with a summation extending over all points of the reciprocal lattice :

$$(8) \quad u_{\underline{k}}(\underline{r}) = \sum_{\underline{m}} \Phi_{\underline{m}} \exp(-i\underline{K}_{\underline{m}}\underline{r})$$

$$V(\underline{r}) = \sum_{\underline{m}} v_{\underline{m}} \exp(-i\underline{K}_{\underline{m}}\underline{r})$$

The Schrödinger equation in K-space is then

$$9- \quad \Phi_{\underline{m}} \left(\frac{\hbar^2 \underline{k}_{\underline{m}}^2}{2\mu} - E \right) + \sum_{\underline{n}} v_{\underline{n}} \Phi_{\underline{n}} \quad \underline{m}-\underline{n}=0$$

with $\underline{k}_{\underline{m}} = \underline{k} - \underline{K}_{\underline{m}}$.

In the case of only one wave the average potential v_0 causes a renormalization of the energy; it is the "work function" in the studies on electrons in metals, but it can also be interpreted in dynamical scattering theory as "average refraction" of the wave.

The simplest case of splitting occurs in the neighborhood of a double degeneracy. Let us suppose that the two waves for which the $E(\underline{k})$ plots intersect in the free-particle approximation are characterized by the reciprocal lattice vectors \underline{K}_0 and \underline{K}_h . It is then reasonable to assume that the Fourier expansion of the function $u_{\underline{k}}(\underline{r})$

in the neighborhood of this point is mainly determined by the contribution of these two waves. Neglecting the effect of the other waves we obtain the equations

$$10- \quad \epsilon_0 \bar{\Phi}_0 - v_{-h} \bar{\Phi}_h = 0$$

$$-v_{h0} \bar{\Phi}_0 + \epsilon_h \bar{\Phi}_h = 0$$

where ϵ_i ($i=0,h$) de-notes the energy change

$$11- \quad \epsilon_i = E - (\hbar^2/2\mu) k_i^2 - v_0$$

For a real interaction potential $V(\underline{r})$ it is $v_{-h} = \frac{v_h}{2}$. Thus the equations (10) lead to the secular equation for the energy values :

$$12- \quad \epsilon_0 \epsilon_h = |v_h|^2$$

From eqs. (10) and (12) we also obtain the relation

$$13- \quad \epsilon_0 |\bar{\Phi}_0|^2 = \epsilon_h |\bar{\Phi}_h|^2$$

which shows that $\bar{\Phi}_h$ goes to zero if ϵ_0 goes to zero, and also that $\epsilon_0 \rightarrow 0$ for $\bar{\Phi}_h \rightarrow 0$. This means that in regions of k -space far away from an intersection point only one wave contributes.

It is not difficult to understand the physical reason for the splitting of the energy levels under the action of the crystal potential. For simplicity let us consider the situation on the k_x -axis at the boundary of the first Brillouin zone of a simple Bravais lattice as in fig. 1.

At this point $k_0^2 = k_1^2 = \pi^2/a^2$. Thus $\epsilon_0 = \epsilon_1 =$ and $|\bar{\Phi}_0| = |\bar{\Phi}_1|$. The secular equation yields $\epsilon = \pm v_1$ (in a simple Bravais lattice a real $V(\underline{r})$ has also real Fourier coefficients since the lattice is invariant with respect to inversions). Equations (10) leads to the result that $\bar{\Phi}_1 = \pm \bar{\Phi}_0$.

The resulting wave functions are therefore given by

$$(14) \quad \psi^{(+)} = \bar{\Phi}_0 \cos(\pi r/a); \quad \psi^{(-)} = \bar{\Phi}_0 \sin(\pi r/a)$$

Since $|\psi|^2$ gives the probability density for the particle, it is seen that $\psi^{(+)}$ leads to a large probability density near the crystal atoms, while $\psi^{(-)}$ goes to zero there.

Thus if the interaction potential $V(r)$ is repulsive (positive), v_0 and v_1 are positive, and the solution which is large at the atomic sites has higher energy than the solution which is small there. In the absence of in-

interaction both solutions and all their linear combinations have the same energy. This explains the original double degeneracy and its removal by the interaction with the lattice. If the atoms of the crystal also absorb radiation then obviously $\psi^{(+)}$ will be attenuated much more than $\psi^{(-)}$ which will penetrate with almost negligible absorption. This effect is called anomalous transmission. It is obvious how this method can be extended to points of higher degeneracy in the original free-particle dispersion relation.

Remark: In our equations entered the mass μ of the propagating particles which is zero in the case of photons. However, this case leads to the wave equation in a medium with periodic dielectric constant. Since this wave equation is invariant with respect to the group, all the considerations above apply to this case too, with the Fourier coefficients of the potential replaced by the Fourier coefficients of the dielectric constant, e_n . For scalar photons (i.e. neglecting the polarization properties of the electromagnetic waves) one obtains the equation⁽⁴⁾

$$15- \quad \Phi_m (k_m^2 - k_v^2) + k_m^2 \sum e_n \Phi_{m-n} = 0$$

Here $k_v = \nu/c$ is the wave number of the photon in vacuum. It plays essentially the same role as the energy E in the method presented above. The dispersion surfaces in X-ray scattering are the surfaces of constant k_v . Eq. (15) can be considered as the photon equivalent of eq. (9).

3. Solution for a Wave Impinging on a Semi-infinite Crystal

In a large but finite crystal the asymptotic solution derived in the preceding section for an infinite crystal is valid at a certain distance from the boundary. Its parameters have to be linked in some way to those of the incident wave field outside the crystal i.e. we have to impose suitable boundary conditions to the asymptotic solution inside the crystal.

To find such boundary conditions one can start with the approximation that the validity of the asymptotic solution extends even to the surface of the crystal. This is certainly not true in a rigorous sense : as we know from the theory of electron bands in finite crystals there are "surface states" at the boundary which die away as one proceeds to the interior atomic layers of the solid. Then, at distances of the order of some hundred Å, the asymptotic solution prevails. On the other hand, in the case of neutrons for example, the interaction between crystal atoms and incident particles takes mainly place at a much larger distance, determined by the mean free path (which, for neutrons, is usually of the order of centimeters), i.e. in a region where the asymptotic solution is already well established. Thus it is likely that boundary conditions which directly link the incident wave and the asymptotic solution will give the right expression for the field inside the crystal, except in the surface region itself.

It is therefore reasonable to impose the boundary conditions of continuity of the wave functions and the currents at the surface of the crystal to a superposition of eigenfunctions of the infinite crystal Hamiltonian H . The continuity of the wave functions implies in particular, for an incident plane wave $\phi_{in} = A \exp(i\hat{k} \cdot r)$, that the tangential (with respect to the surface) component of the wave vector remains unchanged:

$$16- \quad \underline{\hat{k}} - (\underline{\hat{k}} \cdot \underline{n}) \underline{n} = \underline{k} - (\underline{k} \cdot \underline{n}) \underline{n}$$

($\underline{\hat{k}}$ = wave vector of the incident wave , \underline{k} = wave vector inside the crystal , \underline{n} = normal to the surface)

Eq.(16) will be referred to as the condition of continuity of phase (or of phase velocity) at the boundary.

Instead of the condition of the continuity of the current at the surface we can use the much simpler condition that follows from the fact that the energy of the incident beam and of the solution inside the crystal has to be the

same since we are considering elastic coherent scattering only (All other interactions could later be included in this theory as "absorptions"). This conservation of energy requires that the inside solution has a wave vector \underline{k} which lies on the dispersion surface corresponding to the energy of the incident wave :

$$17- \quad E = \hbar^2 \underline{k}^2 / 2$$

It is easy to determine the \underline{k} -vectors graphically from these conditions (fig. 5) : If we draw in the reduced zone scheme the incident vector in such a way that it points at Γ , the center of the zone, and if we draw that normal to the surface which passes through the other end of \underline{k} , then the first condition is satisfied for all vectors which start from that normal \hat{n} and end at Γ . The second condition means that these vectors have to originate at the dispersion surface of energy E . Thus the intersection points of \hat{n} with the dispersion surface determine the wave vectors which contribute to the inside solution. In fig. 5 we have illustrated the case where the dispersion surface has a form typical for the interaction of two free-particle waves characterized by the reciprocal lattice vectors $\underline{K}_0 = \underline{0}$ and \underline{K}_h . As one can see from this figure, there are two \underline{k} -vectors in the first Brillouin zone that contribute to the solution; they are determined by the points A_1 and A_2 .

The solutions corresponding to the different admissible \underline{k} -vectors have to be combined in a way as to satisfy the detailed boundary conditions. Again we limit our discussion to the situation of Fig. 5: To simplify the writing we choose our coordinates in such a way that the boundary corresponds to $z = 0$, and we denote an arbitrary position vector $(x, y, 0)$ in the surface by \underline{s} . Taking into account the fact that the inside solutions are superpositions of waves $\bar{\psi}_0 \exp(i \underline{k} \cdot \underline{r})$

and $\bar{\Phi}_h \exp(i(\underline{k}-\underline{k}_h) \cdot \underline{r})$ we can write the condition of the continuity of the wave functions at the surface in the form

$$18- \quad \phi_{in} \exp(i\underline{k} \cdot \underline{s}) = (\Phi_0^1 + \Phi_0^2) \exp(i\underline{k} \cdot \underline{s}) + (\bar{\Phi}_h^1 + \bar{\Phi}_h^2) \exp(i(\underline{k}-\underline{k}_h) \cdot \underline{s})$$

where the superscripts 1 and 2 refer to the points A_1 and A_2 and where we have used the fact that $\underline{k}_1 \cdot \underline{s} = \underline{k}_2 \cdot \underline{s} = \underline{k} \cdot \underline{s}$. Since equation (18) has to be satisfied identically (for all values of \underline{s}) we have to conclude that

$$19- \quad \phi_{in} = \Phi_0^1 + \Phi_0^2$$

and

$$20- \quad \bar{\Phi}_h^1 = -\bar{\Phi}_h^2$$

From Eq. (10) we have

$$\bar{\Phi}_h^i = (\epsilon_0^i / v_{-h}) \Phi_0^i \quad (i=1,2)$$

so that

$$21- \quad \epsilon_0^2 \Phi_0^2 = -\epsilon_0^1 \Phi_0^1$$

and that therefore

$$22- \quad \phi_{in} = (1 - \epsilon_0^1 / \epsilon_0^2) \Phi_0^1$$

or

$$\Phi_0^1 = \epsilon_0^2 \phi_{in} / (\epsilon_0^2 - \epsilon_0^1)$$

$$\Phi_0^2 = \epsilon_0^1 \phi_{in} / (\epsilon_0^2 - \epsilon_0^1)$$

$$23- \quad \bar{\Phi}_h^1 = \frac{\epsilon_0^1 \epsilon_0^2}{v_{-h} (\epsilon_0^2 - \epsilon_0^1)} \phi_{in}$$

$$\bar{\Phi}_h^2 = \frac{-\epsilon_0^1 \epsilon_0^2}{v_{-h} (\epsilon_0^2 - \epsilon_0^1)} \phi_{in}$$

The case of a wave leaving the crystal is solved in exactly the same manner.

The two waves distinguished by the superscripts 1 and 2 have the same frequency (energy) and their phases are fixed relative to each other by the equations (23).

Since the waves propagate with different wave vectors \underline{k}_1 and \underline{k}_2 , their superposition within the crystal will form a characteristic interference pattern called "Pendellösung" by EWALD in this work on dynamical scattering of X-rays⁽²⁾.

We shall illustrate this oscillatory behaviour of the solution inside the crystal for the particular case in which $\epsilon_0^2 = -\epsilon_0^1$.

This corresponds to the case where the normal \hat{n} passes through a point which is symmetrical with respect to the two branches of the dispersion curves; this means that it passes through the intersection point of the free-particle dispersion curves. Then it follows from the equations (23) that

$$24- \quad \phi_0^1 = \phi_0^2 = \phi_{in}/2$$

and

$$25- \quad \phi_h^1 = -\phi_h^2 = \phi_{in}/2$$

with the abbreviation

$$\eta = \frac{\epsilon_0^1}{v_{-h}}$$

Thus the superposition of the two waves (24) gives

$$26- \quad \begin{aligned} \Phi_0 &= (\phi_{in}/2)\exp(i\underline{k}_1 \cdot \underline{r}) + (\phi_{in}/2)\exp(i\underline{k}_2 \cdot \underline{r}) \\ &= \phi_{in} \cos(\delta \underline{k} \cdot \underline{r}) \exp(i\bar{\underline{k}} \cdot \underline{r}) \end{aligned}$$

where

$$27- \quad \delta \underline{k} = \underline{k}_1 - \underline{k}_2; \quad \bar{\underline{k}} = (\underline{k}_1 + \underline{k}_2)/2$$

$\delta \underline{k}$ is equal to the vector $A_1 A_2$: therefore it points along the normal \hat{n} , i.e. along the z -direction so that $\delta \underline{k} \cdot \underline{r} = cz$, where $c = |\delta \underline{k}|$ is some constant. The intensity $|\Phi_0|^2$ thus has planes of maximum $cz = (m+1/2)\pi$.

In the same way we obtain for the superposition of the waves (25)

$$28- \quad \Phi_h = \phi_{in} \sin(cz) \exp(i\bar{\underline{k}} \cdot \underline{r} - \underline{k}_h \cdot \underline{r})$$

Obviously, the nodal planes of Φ_h coincide with the planes of maximum amplitude of Φ_0 and vice versa.

The superposition of the waves of index 0 and of index h gives rise to still more complicated interference patterns.

However as we have seen: at the planes $z = m\pi/c$ and $z = (m + 1/2)\pi/c$ only one wave is present; therefore the solution simplifies considerably at these points. The direction of the wave emerging from a finite crystal differs for waves of index h from that of waves of index o . The wave of index o corresponds (outside of the crystal) to the direction of the unreflected beam while the wave of index h gives rise on exit to the wave scattered by the plane h . Thus the effect of the pendulum solution is to change periodically (as a function of crystal thickness) the relative intensities of these two emerging beams in such a way that at the corresponding nodal planes one wave is absent.

It is possible to show experimentally this effect by studying the emerging radiation in the case of a transmission through a wedge-shaped crystal.

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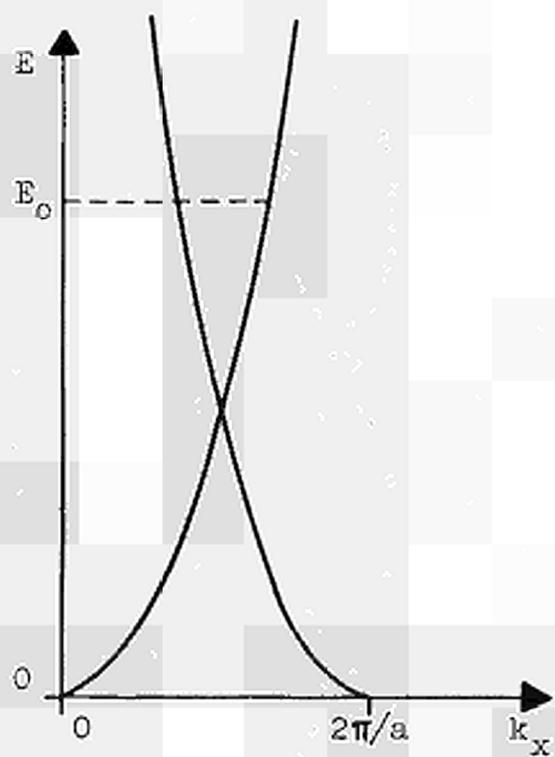


Fig. 1a

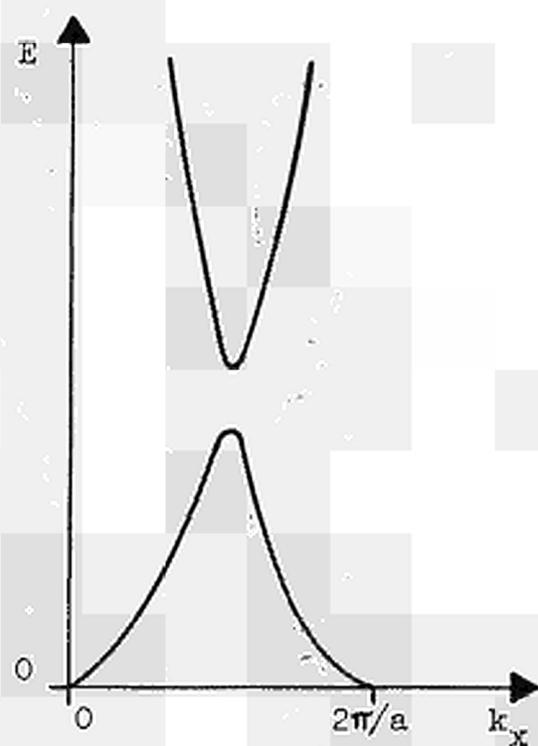


Fig. 1b

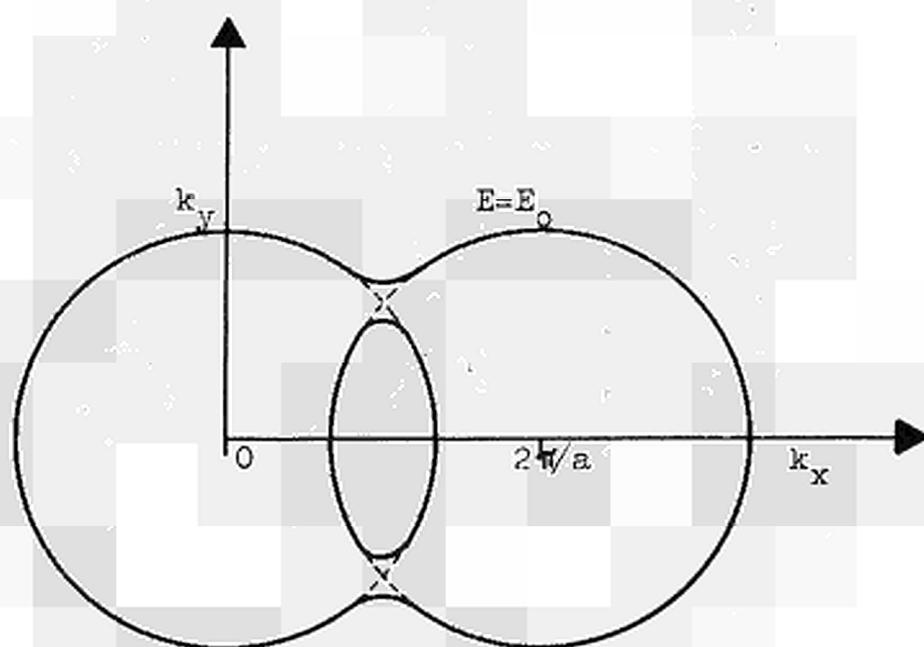


Fig. 2

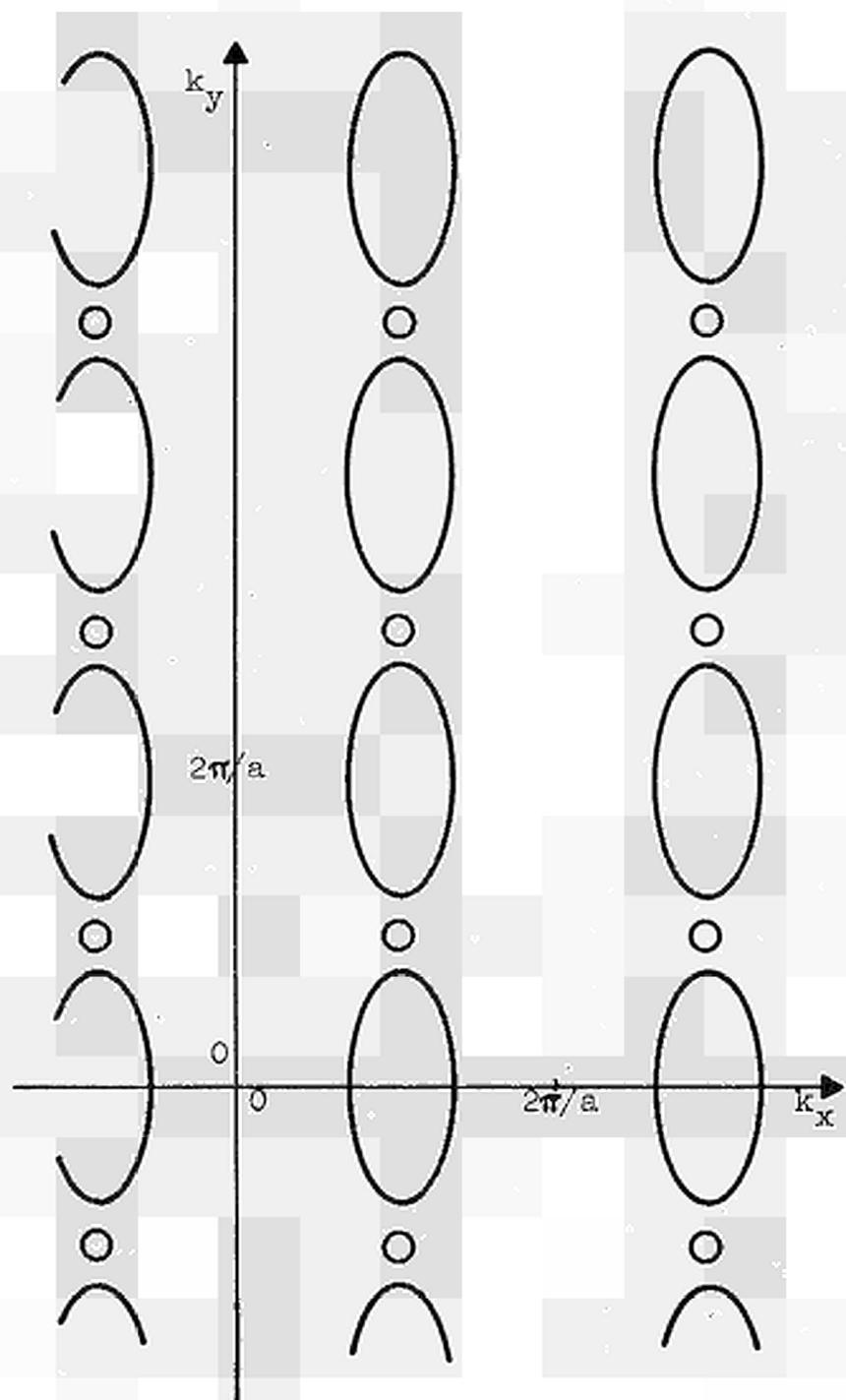


Fig. 3

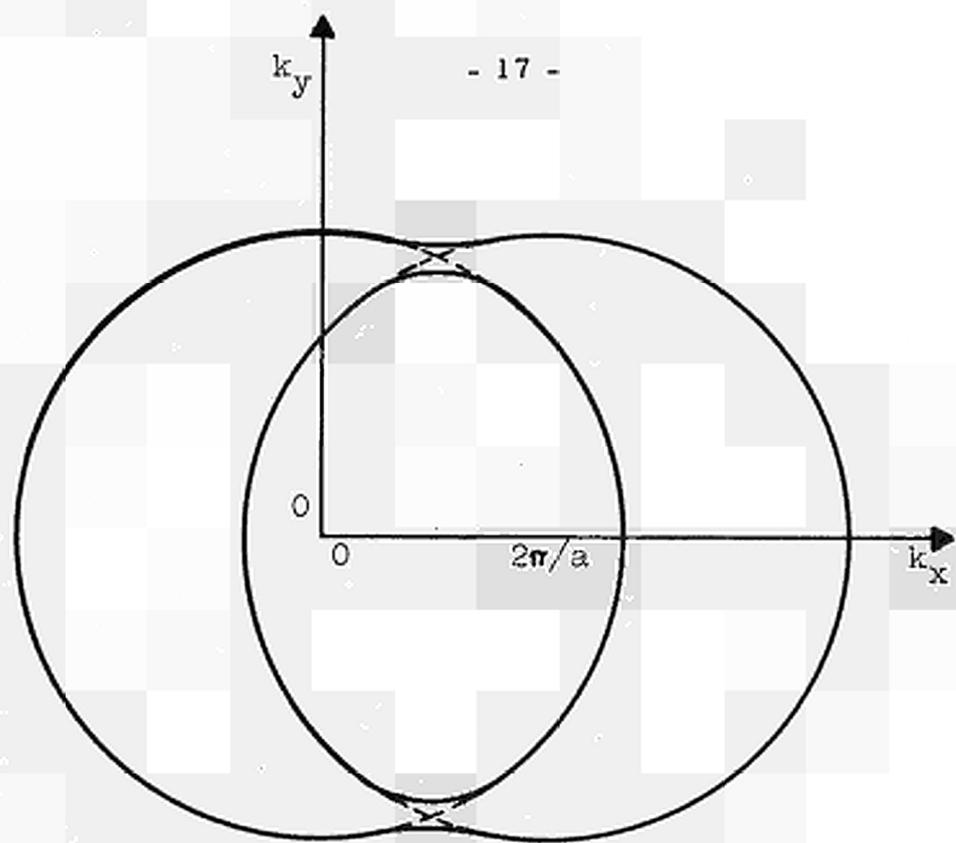


Fig. 4

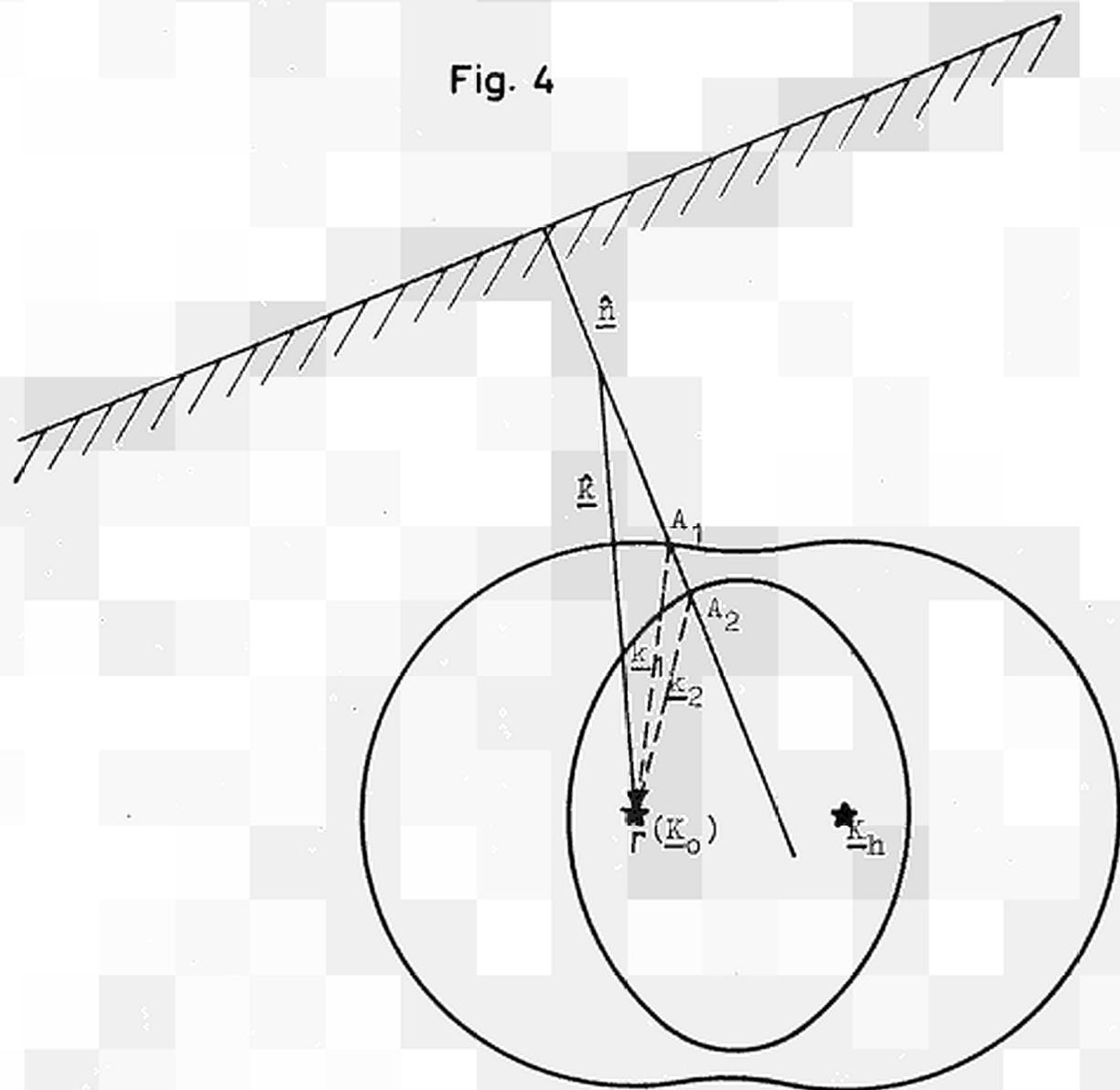


Fig. 5

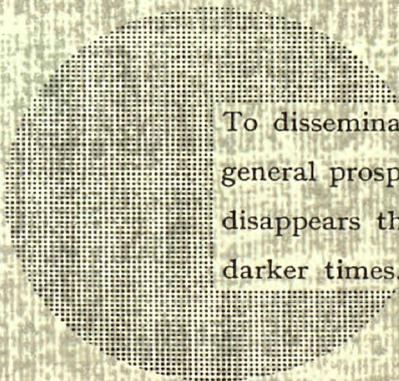
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Alfred Nobel

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