

Collision integral for elastic scattering of electrons and phonons

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The form of the collision integral describing elastic scattering of electrons and phonons by crystal defects is discussed once again. It is shown that in the absence of detailed balance for the transition probability ( $W_{kk'} \neq W_{k'k}$ ), the collision integral does not contain Fermi or Bose factors ( $1 \mp n_k$ ) contrary to the point of view widespread in the literature. It turns out to be independent of the type of particle statistics and to be linear in occupation numbers,  $I_k = \sum_{k'} (W_{kk'} n_{k'} - W_{k'k} n_k)$ . A proof is given of the  $H$ -theorem on the increase in entropy of a system of elastically scattered electrons and phonons.

A system of electrons or phonons scattered elastically by randomly situated static crystal defects is one of the simplest objects in kinetics. However, paradoxical though this may be, in the majority of textbooks and monographs (for example cf. Refs. 1–10) the description of this object is not quite correct. Historically a conviction has arisen that the collision integral in the kinetic equation

$$\frac{dn_k}{dt} = I_k, \tag{1}$$

describing elastic scattering has the form

$$I_k = \sum_{k'} [W_{kk'} n_{k'} (1 \mp n_k) - W_{k'k} n_k (1 \mp n_{k'})], \tag{2}$$

where  $n_k$  is the distribution function, and  $W_{kk'}$ , a characteristic of the scattering centers, is the probability of a transition per unit time from a state with momentum  $k'$  into a state with momentum  $k$ . The upper sign refers to fermions, and the lower one to bosons; for the sake of definiteness we shall refer to the former as electrons and to the latter as phonons. It is accepted that (2) expresses the balance between arrivals and departures taking into account the type of particle statistics. Having written down (2) many authors<sup>1–6</sup> take the following step—they appeal to the principle of detailed balance

$$W_{kk'} = W_{k'k} \tag{3}$$

and reduce the collision integral to the form

$$I_k = \sum_{k'} W_{kk'} (n_{k'} - n_k). \tag{4}$$

At the same time it is asserted that (3) is a universal law stemming from the principle of microreversibility (i.e., from the invariance of the equations of motion under time reversal). As an additional argument frequent use is made of the consideration that (3) guarantees the vanishing of the collision integral in the case of an equilibrium distribution of the particles.

In this note we first of all want to call attention to the fact that the principle of detailed balance does not have the general nature ascribed to it and is fulfilled only under some specific assumptions concerning the properties of the medi-

um. Our second and principal aim is to convince the readers that in the absence of detailed balance expression (2) is erroneous, to give a correct expression for the collision integral and to investigate its principal properties.

The question of the range of applicability of relation (3), generally speaking, has been discussed in the literature.<sup>11–13</sup> It is known both from quantum and from classical mechanics that the invariance of the equations of motion under time reversal leads not to the relation (3) but to the equation

$$W_{kk'} = W_{-k', -k}, \tag{5}$$

often referred to as the reciprocity theorem.<sup>14</sup> Detailed balance holds in the case when (5) is supplemented by the very specific assumption of invariance under spatial inversion  $W_{kk'} = W_{-k, -k'}$ . For crystals without a center of symmetry this assumption does not hold and the principle of detailed balance is not valid<sup>11</sup>. The violation of the principle of detailed balance and the validity of relation (5) can be easily understood from the pictorial considerations of Fig. 1.

The probability of elastic scattering  $W_{kk'}$  satisfies the integral relation

$$\sum_{k'} W_{kk'} = \sum_{k'} W_{k'k}, \tag{6}$$

established by Stukelberg and stemming from the unitarity of the  $S$ -matrix.<sup>16</sup> This condition is sufficient to prove the vanishing of  $I_k$  in thermodynamic equilibrium.

Observable phenomena associated with the absence of detailed balance are now known. Such phenomena are the photogalvanic effect,<sup>15</sup> the anomalous Hall effect,<sup>17</sup> the kinetics of gases with rotational degrees of freedom.<sup>18</sup> Therefore the question of the form of the kinetic equation in the absence of detailed balance is not only of academic, but also of practical interest.<sup>2)</sup>

The erroneous nature of relation (2) is directly shown by

<sup>1)</sup>The absence of detailed balance can be associated also with magnetic effects which violate relation (5).<sup>15</sup>

<sup>2)</sup>In the well-known textbook of Ref. 19 only those physical situations are discussed for which  $W_{kk'} = W_{k'k}$ .

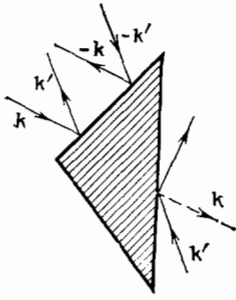


FIG. 1.

the nonphysical nature of its consequences. In order to see this we consider the case of a classical wave field, i.e., the case of Bose-statistics in the limit  $n \gg 1$ . In the classical limit (2) gives

$$I_{\mathbf{k}} = n_{\mathbf{k}} \sum_{\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'}^a n_{\mathbf{k}'}, \quad W_{\mathbf{k}\mathbf{k}'}^a = W_{\mathbf{k}\mathbf{k}'} - W_{\mathbf{k}'\mathbf{k}}. \quad (7)$$

Contrary to common sense the scattering turns out to be completely asymmetric and nonlinear. In fact (7) admits that it is possible to obtain a nonlinear equation for the intensities from classical equations of motion that are linear in the wave amplitudes. It can be easily shown that collisions described by (7) do not increase the entropy of the system of waves; they should lead to the formation of singularities in the distribution of waves in  $\mathbf{k}$ -space and to sharp anomalies in the optical and thermal properties of crystals.

The error made in writing (2) consists in the unjustified generalization of results obtained in the first Born approximation to higher orders of perturbation theory in the defect potential. In the first Born approximation one has independently of the symmetry properties of the scattering center  $W_{\mathbf{k}\mathbf{k}'} = W_{\mathbf{k}'\mathbf{k}}$ <sup>12,14</sup> so that the nonlinear contributions to (2) cancel automatically. In higher orders of perturbation theory for  $W_{\mathbf{k}\mathbf{k}'}$ , when detailed balance is absent summations over intermediate states arise. Thus, in the second Born approximation we have

$$W_{\mathbf{k}\mathbf{k}'} = 2\pi N_0 \delta(\omega - \omega') \left( |V_{\mathbf{k}\mathbf{k}'}|^2 + 2 \operatorname{Re} \sum_{\mathbf{k}''} \frac{V_{\mathbf{k}\mathbf{k}''} V_{\mathbf{k}''\mathbf{k}'} V_{\mathbf{k}'\mathbf{k}}}{\omega - \omega'' + i0} \right), \quad (8)$$

where  $N_0$  is the defect concentration,  $V_{\mathbf{k}\mathbf{k}'} = V_{\mathbf{k}'\mathbf{k}}^*$  is the matrix element of the perturbation for an individual center,  $\omega_{\mathbf{k}}$  is the dispersion law (energy) of the quasiparticles.<sup>3)</sup> The Born series for  $W_{\mathbf{k}\mathbf{k}'}$  does not contain occupation numbers for intermediate states. However, it is quite clear that in investigating the kinetic properties of a many-particle system it is just as necessary to take them into account as to take into account the occupation numbers for the initial and final states. Consequently, use of expression (2) outside the framework of the first Born approximation is not correct. In order to obtain the collision integral in the absence of detailed balance it is necessary to use the consistent procedure of the derivation of the kinetic equation which is explicitly based on the apparatus of second quantization.

<sup>3)</sup>We are using the system of units in which  $\hbar = 1$ .

It is remarkable that the final result turns out to be universal and simple. Both for the electrons and for the phonons the collision integral is of the form (4) where  $W_{\mathbf{k}\mathbf{k}'} \neq W_{\mathbf{k}'\mathbf{k}}$  is the exact probability of scattering. It coincides with the collision integral for a classical gas of particles or a nondegenerate electron gas. It can be said that the cancellation of the contributions to  $I_{\mathbf{k}}$  nonlinear in  $n_{\mathbf{k}}$  associated with the "arrival" and "departure" occurs not only in the first Born approximation, but also in the higher orders of perturbation theory. Taking (6) into account the kinetic equation for the elastic scattering can be written finally in the form

$$\frac{dn_{\mathbf{k}}}{dt} = \sum_{\mathbf{k}'} (W_{\mathbf{k}\mathbf{k}'} n_{\mathbf{k}'} - W_{\mathbf{k}'\mathbf{k}} n_{\mathbf{k}}). \quad (9)$$

Apparently the first to give the correct expression for the collision integral were Kohn and Luttinger.<sup>20</sup> Their method of derivation was quite cumbersome. But in fact the final result (9) is very transparent; it can be discerned already from the form of the initial dynamic equations of motion. Let us consider the Hamiltonian of the interaction responsible for elastic scattering. In the representation of second quantization we have

$$\hat{V} = \sum_{\mathbf{k}_1 \mathbf{k}_2} V_{\mathbf{k}_1 \mathbf{k}_2} a_{\mathbf{k}_1}^+ a_{\mathbf{k}_2}, \quad (10)$$

where  $a_{\mathbf{k}}^+$  and  $a_{\mathbf{k}}$  are particle creation and annihilation operators. It can be easily seen that independently of the type of particle statistics (i.e., of the type of commutation relations for  $a^+$  and  $a$ ) we have the commutators

$$[\hat{V}, a_{\mathbf{k}}^+ a_{\mathbf{k}'}] = \sum_{\mathbf{k}_1} (V_{\mathbf{k}_1 \mathbf{k}} a_{\mathbf{k}_1}^+ a_{\mathbf{k}'} - V_{\mathbf{k}' \mathbf{k}_1} a_{\mathbf{k}}^+ a_{\mathbf{k}_1}). \quad (11)$$

Thus, in agreement with the rules of quantum mechanics,<sup>14</sup> the equations of motion for the averages  $\langle a_{\mathbf{k}}^+ a_{\mathbf{k}'} \rangle$  turn out to be the same for electrons and for phonons and are linear;

$$\left[ \frac{\partial}{\partial t} + i(\omega_{\mathbf{k}'} - \omega_{\mathbf{k}}) \right] \langle a_{\mathbf{k}}^+ a_{\mathbf{k}'} \rangle = i \sum_{\mathbf{k}_1} (V_{\mathbf{k}_1 \mathbf{k}} \langle a_{\mathbf{k}_1}^+ a_{\mathbf{k}'} \rangle - V_{\mathbf{k}' \mathbf{k}_1} \langle a_{\mathbf{k}}^+ a_{\mathbf{k}_1} \rangle). \quad (12)$$

The relation (12) also describes the evolution of a classical wave field if we interpret  $a_{\mathbf{k}}$  as the normal amplitude of a wave satisfying the Hamiltonian equation of motion<sup>21</sup>

$$\frac{\partial a_{\mathbf{k}}}{\partial t} + i\omega_{\mathbf{k}} a_{\mathbf{k}} = -i \frac{\delta V \{a^*, a\}}{\delta a_{\mathbf{k}}^*} \quad (13)$$

(the notation  $\langle \dots \rangle$  here denotes averaging over the ensemble of waves).

The subsequent procedure of obtaining from (12) the kinetic equation for the distribution function  $n_{\mathbf{k}} = \langle a_{\mathbf{k}}^+ a_{\mathbf{k}} \rangle$  generally does not contain any elements which depend on the type of particle statistics, it is completely analogous to the procedure used in Ref. 19 to obtain the classical kinetic equation. We must write the Bogolyubov chain of equations for the averages (in the classical case the ensemble averages). The first equation of this chain relates  $n_{\mathbf{k}}$  to the average  $V_{\mathbf{k}\mathbf{k}_1} \langle a_{\mathbf{k}}^+ a_{\mathbf{k}_1} \rangle$ , the second expresses this average in terms of a quantity of a higher order in  $V$ , etc. At the last step the number of which depends on the required accuracy of taking the

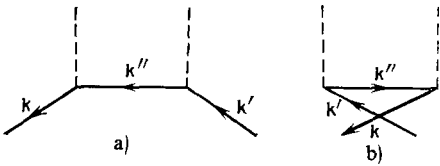


FIG. 2.

interaction into account we break the chain by neglecting the correlations and assuming  $\langle a_{k_i}^+ a_{k_j} \rangle = \delta_{k_i, k_j} n_{k_i}$ . From (12) it is abundantly clear that the kinetic equation that is obtained must be linear in  $n_k$ ; consequently it coincides with equation (9) the validity of which for describing the kinetic property of a nondegenerate gas of particles is obvious. The probability  $W_{kk'}$  is the usual Born series in powers of  $V_{kk'}$ . The criterion for the applicability of the kinetic equation is the usual inequality  $\omega_k \gg \gamma_k$ , where  $\gamma_k$  is the frequency of electron-impurity or phonon-impurity collisions.

It should be said that in the second Born approximation it is not difficult to obtain equation (9) by the traditional method based on representing the collision integral in the form of the difference between the rates of arrival of particles into the state  $|k\rangle$  and departure from it.<sup>4)</sup> It is of interest to trace how in this case the cancellation of nonlinear terms occurs. For the sake of simplicity we restrict ourselves to the case of Bose statistics. The contribution to  $I_k$  associated with arrival processes can be written with an accuracy to terms of order  $V^3$  in the form

$$I_k^{\text{arr}} = 2\pi N_0 \sum_{k'} \delta(\omega - \omega') \left| \langle n' - 1, n + 1 | V | n', n \rangle + \sum_s \frac{\langle n' - 1, n + 1 | V | s \rangle \langle s | V | n', n \rangle}{E - E_s + i0} \right|^2. \quad (14)$$

This formula can be derived rigorously in scattering theory,<sup>23</sup> according to which the probability of a transition per unit time from the state  $a$  to the state  $b$  can be expressed in terms of the square of the modulus of the matrix element of the  $\hat{T}$  operator which can be represented in the form of a series in powers of  $\hat{V}$ :

$$W_{ba} = 2\pi N_0 |\hat{T}_{ba}|^2 \delta(E_a - E_b), \quad (15)$$

$$\hat{T} = \hat{V} + \hat{V} (E - H_0 + i0)^{-1} \hat{V} + \dots;$$

here  $H_0 = \sum_k \omega_k a_k^+ a_k$  is the unperturbed Hamiltonian for the system.

Two types of intermediate states  $|s\rangle$  are possible. The states of the first kind  $|s\rangle = |n'' + 1, n' - 1\rangle$ , correspond to the graph a) in Fig. 2. For them we have  $E - E_s = \omega - \omega''$ . The states of the second type,  $|s\rangle = |n'' - 1, n + 1\rangle$ , correspond to the graph b) in Fig. 2 and to the energy difference  $E - E_s = \omega'' - \omega$ . Using the explicit expression (10) for  $\hat{V}$  we transform the square of the modulus appearing (14) to the form

$$n'(n+1) \left\{ |V_{kk'}|^2 + 2 \operatorname{Re} \sum_{k''} \left[ \frac{n''}{\omega'' - \omega + i0} + \frac{n'' + 1}{\omega - \omega'' + i0} \right] \times V_{kk''} V_{k''k'} V_{k'k} \right\}. \quad (16)$$

<sup>4)</sup>And also with the aid of Keldysh's diagram technique for nonequilibrium processes.<sup>19</sup>

Taking into account the Hermitian nature of  $\hat{V}$  and the well-known identity  $(x + i0)^{-1} = (P/x) - i\pi\delta(x)$ , where  $P$  is the symbol for the principal value, one can easily demonstrate the cancellation of the contributions to  $I_k^{\text{arr}}$  cubic in  $n_k$ . Finally we obtain

$$I_k^{\text{arr}} = \sum_{k'} n_{k'} (1 + n_k) W_{kk'}, \quad (17)$$

where  $W_{kk'}$  is given by formula (8).

The rate of departure of particles  $I_k^{\text{dep}}$  can be calculated in a similar manner. As is quite evident it is described by (14) if we interchange the subscripts  $k$  and  $k'$  in the square of the modulus (16) appearing in it. The final expression for  $I_k^{\text{dep}}$  is the following:

$$I_k^{\text{dep}} = \sum_{k'} n_k (1 + n_{k'}) W_{k'k}; \quad (18)$$

it does not coincide with the expression for  $I_k^{\text{dep}}$  given by formula (2).

In the end the terms quadratic in  $n_k$  also cancel and we again arrive at equations (4) and (9). The fact that  $I_k^{\text{dep}}$  contains the probability  $W_{kk'}$ , and not  $W_{k'k}$ , is associated with the presence in the figure brackets (16) of contributions containing  $\delta(\omega - \omega'')$ . On the one hand these contributions determine the antisymmetric probability of scattering  $W_{kk'}^a$ ,<sup>13</sup> and on the other hand they change sign when the replacement  $k \leftrightarrow k'$  is made. If there is a center of symmetry then  $V_{kk'} = V_{-k-k'}$  and these contributions vanish.

It is useful to note that dividing the collision integral (9) into an incoming and outgoing terms (17), (18) is for  $n_k \gtrsim 1$  quite arbitrary, and the nonlinearity of the quantities  $I_k^{\text{arr, dep}}$  in terms of the occupation numbers can not be manifested in any kinetic effect.

We make two generalizations. If inelasticity of scattering (scattering of electrons by phonons) is taken into account the conclusion that in  $I_k$  there are no terms nonlinear in  $n_k$  is no longer valid. However, also in this case formula (2) remains valid only in the first Born approximation. In higher orders of perturbation theory contributions appear in  $I_k$  containing  $n^3$ ,  $n^4$ , etc. A similar conclusion can be drawn also in the case of pair interaction of particles described by a Hamiltonian of the form  $a_1^+ a_2^+ a_3 a_4$  and here the well-known expression for the collision integral

$$I_k = \sum_{k_1 k_2} W_{kk'; k_1 k_2} \times [(1 \mp n)(1 \mp n') n_1 n_2 - (1 \mp n_1)(1 \mp n_2) n n'],$$

is valid, generally speaking, only in the first Born approximation, when  $W_{kk'; k_1 k_2} = W_{k_1 k_2; kk'}$ .

We consider some general properties of the kinetic equation (9). Using the relation (6) it can be easily shown that it guarantees the evolution of the electron distribution functions  $n_k$  which is compatible with the Pauli principle—if at  $t = 0$  the inequality  $n_k \leq 1$  holds, then it remains valid also at all subsequent instants of time.

We now prove for equation (9) the Boltzmann  $H$ -theorem on the increase of the entropy of the system of electrons and phonons. The entropy of nonequilibrium Fermi and Bose gases is defined by the formulas<sup>24</sup>

$$S_F = - \sum_{\mathbf{k}} [n_{\mathbf{k}} \ln n_{\mathbf{k}} + (1 - n_{\mathbf{k}}) \ln (1 - n_{\mathbf{k}})],$$

$$S_B = - \sum_{\mathbf{k}} [n_{\mathbf{k}} \ln n_{\mathbf{k}} - (1 + n_{\mathbf{k}}) \ln (1 + n_{\mathbf{k}})].$$

In the classical limit, i.e., for  $n_{\mathbf{k}} \ll 1$  for electrons and for  $n_{\mathbf{k}} \gg 1$  for phonons, these expressions reduce to

$$S_F^0 = - \sum_{\mathbf{k}} n_{\mathbf{k}} \ln n_{\mathbf{k}},$$

$$S_B^0 = \sum_{\mathbf{k}} \ln n_{\mathbf{k}}.$$

We now show that (9) leads to such an evolution of the system for which  $\dot{S} > 0$ , with the equality holding only when the equilibrium distribution  $n(\omega_{\mathbf{k}}) = \bar{n}_{\mathbf{k}}(t=0)$  is attained; the bar means averaging over the equal frequency surface. We begin the proof starting with the case of a nondegenerate electron gas. According to (9) and (20), we have

$$\dot{S}_F^0 = \frac{1}{2} \sum_{\mathbf{k}\mathbf{k}'} (W_{\mathbf{k}'\mathbf{k}} n_{\mathbf{k}} - W_{\mathbf{k}\mathbf{k}'} n_{\mathbf{k}'}) \ln \frac{n_{\mathbf{k}}}{n_{\mathbf{k}'}}. \quad (21)$$

In the absence of detailed balance some of the terms in the sum can be negative, and therefore the  $H$ -theorem is not obvious. We carry out in (21) the replacement

$$n_{\mathbf{k}} = \bar{n}(\omega_{\mathbf{k}}) e^{x_{\mathbf{k}}}. \quad (22)$$

Since  $W_{\mathbf{k}\mathbf{k}'} \propto \delta(\omega - \omega')$ , equilibrium corresponds to  $x_{\mathbf{k}} = 0$ . Taking (6) into account we first of all show that  $\dot{S}_F^0(x=0) = 0$ , while for small deviations from equilibrium we have ( $|x_{\mathbf{k}}| \ll 1$ )  $\dot{S}_F^0 > 0$ . Then we show that the identity

$$\sum_{\mathbf{k}} \frac{\partial \dot{S}_F^0}{\partial x_{\mathbf{k}}} = \dot{S}_F^0. \quad (23)$$

holds. From this it follows that the extrema of  $\dot{S}_F^0(x_{\mathbf{k}})$  can be attained only at those points where  $\dot{S}_F^0 = 0$ . However, if we admit the possibility that the function  $\dot{S}_F^0(x)$  can change sign, then it follows that if it is positive in the neighborhood of zero then an extremum (saddle point) exists for which  $\dot{S}_F^0 > 0$ . We have arrived at a contradiction. Consequently,  $\dot{S}_F^0 > 0$  for any deviation from equilibrium.

As can be seen from (9), (19) and (20), the quantity  $\dot{S}_F$  can be written in the form  $\dot{S}_F^0(n_{\mathbf{k}}) + \dot{S}_F^0(1 - n_{\mathbf{k}})$ . Since above we have shown that  $\dot{S}_F^0$  is formally nonnegative for any  $n_{\mathbf{k}}$ , then the fact that the  $H$ -theorem holds for Fermi particles becomes obvious.

We now go to Bose particles. We consider

$$\dot{S}_B^0 = \sum_{\mathbf{k}\mathbf{k}'} W_{\mathbf{k}\mathbf{k}'} (e^{x_{\mathbf{k}}} - x_{\mathbf{k}'} - 1). \quad (24)$$

The validity of the  $H$ -theorem for small deviations from equilibrium can again be verified directly. In order to prove it for finite deviations we take into account the identity

$$\sum_{\mathbf{k}} \frac{\partial \dot{S}_B^0}{\partial x_{\mathbf{k}}} = 0. \quad (25)$$

We introduce the function  $R(x) = \dot{S}_B^0 - \lambda \dot{S}_F^0$ , where  $\lambda < 1$  is a positive parameter. For  $x_{\mathbf{k}} = 0$   $R = 0$ , and for  $|x_{\mathbf{k}}| \ll 1$   $R > 0$ . Taking (23) and (25) into account we have

$$\sum_{\mathbf{k}} \frac{\partial R}{\partial x_{\mathbf{k}}} = -\lambda \dot{S}_F^0 \leq 0. \quad (26)$$

Consequently  $R(x)$  does not have any extrema other than a minimum at zero, and is a non-negative quantity. From this follows the fact that  $\dot{S}_B^0(x)$  is positive.

For arbitrary occupation numbers we can formally represent  $\dot{S}_B(n_{\mathbf{k}})$  in the form

$$\dot{S}_B(n_{\mathbf{k}}) = \dot{S}_F^0(n_{\mathbf{k}}) - \dot{S}_F^0(1 + n_{\mathbf{k}}). \quad (27)$$

Taking into account the identity

$$\frac{d}{d\xi} \dot{S}_F^0(n_{\mathbf{k}} + \xi) = -\dot{S}_B^0(n_{\mathbf{k}} + \xi), \quad (28)$$

in which  $\xi$  is an arbitrary parameter, we have  $\dot{S}_B^0(n_{\mathbf{k}}) \geq 0$ . Thus, the  $H$ -theorem has been proved for all cases of elastic scattering.

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