



# Negative density of states: Screening, Einstein relation, and negative diffusion

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(Received 26 August 2008; published 30 October 2008)

In strongly interacting electron systems with low density the thermodynamic density of states is negative at low temperatures. This creates difficulties with understanding of the Einstein relation between conductivity and diffusion coefficient. Using the expression for electrochemical potential that takes into account the long-range part of the Coulomb interaction it is shown that at negative density of states the Einstein relation gives a negative sign of the diffusion coefficient  $D$ , but under this condition there is no thermodynamic limitation on the sign of  $D$ . That happens because the unipolar relaxation of inhomogeneous electron density is not described by the diffusion equation. The relaxation goes much faster due to electric forces caused by the inhomogeneous electron density. The diffusion coefficient is irrelevant in this case and it is not necessarily positive because the diffusion process does not contribute to the positive production of entropy. In the case of bipolar diffusion, negative  $D$  results in a global absolute instability that leads to formation of neutral excitons. Graphene is considered as an example of a system where the density relaxation is expected to be due to electric force rather than diffusion. It may also have a negative density of states.

DOI: [10.1103/PhysRevB.78.155130](https://doi.org/10.1103/PhysRevB.78.155130)

PACS number(s): 71.27.+a, 71.35.Ee, 78.67.De

## I. INTRODUCTION

The idea of the Einstein relation was put forward by Einstein<sup>1</sup> and von Smoluchowsky<sup>2</sup> in 1905–1906. Both scientists considered Brownian motion in the presence of gravitational force. The result is the relation between mobility  $u$  in the field and the diffusion coefficient  $D$ . In case of electric field and particles with charge  $e$  the Einstein relation has a form

$$eD = Tu, \quad (1)$$

where  $T$  is the temperature in energy units. The main idea was equivalence of an external force and a density gradient. Of course, neither Einstein<sup>1</sup> nor von Smoluchowsky<sup>2</sup> cared about negligible mutual gravitational or any other small interaction of the Brownian particles. However, the interaction might be important for electron systems.

The formulation of the Einstein relation for electrons is based on the electrochemical potential, the thermodynamic function that, like temperature and pressure, should be the same at all points of the system in the equilibrium state. The usual arguments are as follows. If an external potential  $\psi$  is applied to the system, the condition of thermodynamic equilibrium reads

$$E_{\text{ec}} = \mu(n) + e\psi = \text{const}, \quad (2)$$

where  $\mu(n)$  is the chemical potential as a function of inhomogeneous electron density  $n$ . In the equilibrium both  $n$  and  $\psi$  are function of coordinates while  $E_{\text{ec}}$  is constant. The temperature  $T$  should also be constant. Therefore, the electrical current density  $\mathbf{j}$  at constant  $T$  can be written in a form<sup>3</sup>

$$\mathbf{j} = -\frac{\sigma}{e} \nabla E_{\text{ec}} = \sigma \mathbf{E} - D \nabla n, \quad (3)$$

where  $\sigma$  is conductivity and  $\mathbf{E} = -\nabla\psi$ . Then one gets relation connecting  $\sigma$  and  $D$ ,

$$\frac{\sigma}{e^2} \frac{d\mu}{dn} = D, \quad (4)$$

which is more general form of the Einstein relation. The derivative  $dn/d\mu$  is called the thermodynamic density of states. For the Boltzmann gas  $d\mu/dn = T/n$  and one gets Eq. (1) if  $\sigma = enu$ . It appears that the derivation of Eq. (4) is independent of the properties of the system and this equation can be considered as a general thermodynamic law.

A simple observation shows however that in the case of nonideal electron gas the Einstein relation needs some comments. We discuss an electron gas in a positive uniform background at low temperatures and low densities when dimensionless parameter  $r_s$  is not very small. Here  $r_s^3 = 3/(4\pi n a_B^3)$  in three-dimensional (3D) case and  $r_s^2 = 1/\pi n_2 a_B^2$  in two-dimensional (2D) case, where  $n$  and  $n_2$  are three- and two-dimensional electron densities, respectively,  $a_B = \hbar^2 \kappa / me^2$  is the Bohr radius,  $m$  is an effective electronic mass, and  $\kappa$  is an effective permittivity.

The problems of dynamic screening and diffusion in a slightly nonideal electron gas ( $r_s \ll 1$ ) with electron-electron interaction were considered in detail about 20 years ago (see Refs. 4–6). In this case the thermodynamic density of states is large and positive. I concentrate here on the strongly nonideal case  $r_s \geq 1$ .

An electron gas on the positive background at low temperatures and low densities has energy  $E$  of the order of  $-e^2 n^{1/d} N / \kappa$ , where  $d=2,3$  is the space dimensionality,  $n$  is the density per unit area or volume, and  $N$  is total number of electrons. Then  $\mu \sim -e^2 n^{1/d} / \kappa$  and  $E$ ,  $\mu$ , and  $d\mu/dn$  are negative.<sup>7,8</sup> The first experimental confirmation of this idea was done by Kravchenko *et al.*,<sup>9,10</sup> but direct quantitative study of this effect was performed by Eisenstein *et al.*<sup>11,12</sup>

The thermodynamic density of states is the compressibility of the electron gas. It has to be positive due to the thermodynamical condition of stability. However, this principle cannot be applied to the charged systems, such as electron

gas, because part of their energy is outside the system in a form of the energy of an electric field. On the other hand, in the case of neutral electron-hole plasma, the situation of negative compressibility can arise leading to collapse of the system. Such a situation is considered at the end of Sec. III.

It follows from Eq. (4) that if  $d\mu/dn$  is negative, the diffusion coefficient  $D$  and conductivity  $\sigma$  have opposite signs. This observation needs an explanation because near the thermodynamic equilibrium both of them have to be positive to provide a positive entropy production due to the Joule heat and due to the relaxation of inhomogeneous density.

## II. ELECTROCHEMICAL POTENTIAL AND STATIC SCREENING

To resolve this contradiction one should include the long-range part of the Coulomb potential created by inhomogeneous electron gas into the function  $E_{\text{ec}}$  in Eq. (2). This contribution is a functional of  $n(\mathbf{r})$ .

To find  $E_{\text{ec}}$ , taking into account electron-electron interaction, one should minimize the Helmholtz energy  $F$  with respect to electron density  $n(\mathbf{r})$  at a given value of  $T$  and  $N$ . For low  $T$  one gets

$$F = \frac{e^2}{2\kappa} \iint \frac{n'(\mathbf{r})n'(\mathbf{r}')d^3rd^3r'}{|\mathbf{r}-\mathbf{r}'|} + \int f(n+n')d^3r + \int en'(\mathbf{r})\psi d^3r - E_{\text{ec}} \int n'(\mathbf{r})d^3r, \quad (5)$$

where  $f$  is the Helmholtz energy density of a homogeneous electron system in the positive background that results from the interaction. Since this interaction comes mainly from the nearest neighbors and  $n(\mathbf{r})$  is a smooth function, one may assume that both  $f$  and the chemical potential  $\mu=df/dn$  are local functions of  $n(\mathbf{r})$ . We assume also that  $n(\mathbf{r})=n+n'(\mathbf{r})$ , where  $n$  is average density and  $n' \ll n$ .

Minimization of this expression with respect to  $n'$  gives the equation

$$E_{\text{ec}} = \mu(n) + e\psi + \frac{d\mu}{dn}n' + \frac{e^2}{\kappa} \int \frac{n'(\mathbf{r}')d^3r'}{|\mathbf{r}-\mathbf{r}'|}. \quad (6)$$

Equation (6) differs from Eq. (2) by the potential of electrons in the right-hand side. Note that this potential is due to the violation of neutrality in a scale much larger than the average distance between electrons. To check this equation we consider thermodynamic equilibrium and find equations for the Thomas-Fermi static screening in three- and two-dimensional cases. Since  $E_{\text{ec}}$  is independent of  $\mathbf{r}$  in thermodynamic equilibrium one may take  $E_{\text{ec}}-\mu(n)$  as a reference point for the total potential  $\varphi$  defined as

$$\varphi = \psi + \frac{e}{\kappa} \int \frac{n'(\mathbf{r}')d^3r'}{|\mathbf{r}-\mathbf{r}'|}. \quad (7)$$

It follows from Eq. (6) that

$$e\varphi = -\frac{d\mu}{dn}n'. \quad (8)$$

The Poisson equation has the form

$$\nabla^2\varphi = -\frac{4\pi(en' - \rho_{\text{ext}})}{\kappa}, \quad (9)$$

where  $\rho_{\text{ext}}$  is density of the external charge. Using Eq. (8) one gets the final equation for the 3D linear screening,

$$\nabla^2\varphi = -q_3^2\varphi - \frac{4\pi\rho_{\text{ext}}}{\kappa}. \quad (10)$$

Here,

$$q_3^2 = \frac{4\pi e^2}{\kappa} \frac{dn}{d\mu} \quad (11)$$

is the reciprocal three-dimensional screening radius.

Consider now a thin layer ( $x$ - $y$  plane) with a 2D electron gas separating two media with dielectric constants  $\kappa_1$  and  $\kappa_2$ . In this case one should substitute  $n \Rightarrow n_2\delta(z)$  and  $\kappa \Rightarrow \bar{\kappa} = (\kappa_1 + \kappa_2)/2$ . The result is<sup>13</sup>

$$\nabla^2\varphi = -q_2\varphi\delta(z) - \frac{4\pi\rho_{\text{ext}}}{\bar{\kappa}}, \quad (12)$$

where

$$q_2 = \frac{2\pi e^2}{\bar{\kappa}} \frac{dn_2}{d\mu}. \quad (13)$$

It is important that Eqs. (10) and (12) are applicable only if the screening is linear ( $n' \ll n$ ).<sup>14</sup> There is another serious problem of applicability of the Thomas-Fermi approximation in the case of negative density of states. Indeed, the dielectric permittivity in this approximation has forms

$$\epsilon(q) = \kappa \left( 1 - \frac{|q_3^2|}{q^2} \right) \quad (14)$$

in the 3D case and

$$\epsilon(q) = \kappa \left( 1 - \frac{|q_2|}{q} \right) \quad (15)$$

in the 2D case. In both cases  $\epsilon(q)$  has roots at  $q=|q_3|, |q_2|$ . The expression for the screened potential  $\varphi$  has the form

$$\varphi(\mathbf{r}) = \int \frac{\varphi_0(\mathbf{q})\exp(i\mathbf{q} \cdot \mathbf{r})d\mathbf{q}}{\epsilon(\mathbf{q})}, \quad (16)$$

where  $\varphi_0$  is a bare potential. Thus, the roots of  $\epsilon$  transform into the first-order poles of the integrand without any reasonable rule of handling those poles. Such a rule follows from the causality for the  $\omega$  plane but not for the  $q$  plane. Moreover, the electrostatic potential should be real and so one cannot add a small imaginary part in the denominator. Therefore it seems that the poles do not have any physical sense.

The reason is that the negative sign of the density of states appears when  $q_3, q_2$  are of the order of the average distance between electrons  $\bar{r}$ . At such distances the very concept of macroscopic field does not have any sense. However, if the bare potential has only harmonics with  $q \ll |q_3|, |q_2|$ , Eqs. (10) and (12) have meaning.

Consider, for example, the screening of a positive charge  $Z$  at a distance  $z_0$  from the plane with 2D gas (plane  $z=0$ ). The solution of Eq. (12) has a form<sup>13</sup>

$$\varphi(\rho) = \int_0^\infty \frac{Z \exp(-qz_0)}{\kappa(q+q_2)} q J_0(q\rho) dq, \quad (17)$$

where  $\rho$  is a polar radius in the plane  $z=0$ . Suppose that  $|q_2|z_0 \gg 1$ . Then the contribution to integral [Eq. (17)] from  $q \approx |q_2|$  is exponentially small and one can ignore  $q$  in the denominator. Then

$$\varphi(\rho) = \frac{Zz_0}{\kappa q_2 (z_0^2 + \rho^2)^{3/2}}. \quad (18)$$

Note that a positive charge creates a small *negative* potential in the plane with electrons if  $q_2 < 0$ . This can be called “overscreening.”

The extra electron density, as calculated from Eq. (8), is

$$en' = -\frac{Zz_0}{2\pi(z_0^2 + \rho^2)^{3/2}}. \quad (19)$$

It is negative and independent of the sign of  $q_2$ . One can see that the total charge

$$\int_0^\infty en' 2\pi\rho d\rho = -Z. \quad (20)$$

Due to the geometry of the problem the electric field is zero below the plane with electrons. As follows from Eq. (8), the signs of charge density and potential are opposite if the density of states is negative.

For the case of two such planes (double quantum well structure) Luryi<sup>15</sup> predicted a small penetration of electric field through the first plane. He considered the case of positive density of states. Then the small penetrating field between two planes has the same direction as the incident field.

Eisenstein *et al.*<sup>11</sup> studied this effect experimentally and found out that at negative density of states the propagating field is opposite to the incident field and this is also a result of the overscreening (see the quantitative theory in Refs. 12, 16, and 17). Negative density of states was also used<sup>18</sup> for the explanation of magnetocapacitance data by Smith *et al.*<sup>19</sup>

### III. CONDUCTIVITY VERSUS DIFFUSION

Now we come back to the problem of negative diffusion. If the system is not in equilibrium, the electric current can be written in the same form as Eq. (3),

$$\mathbf{j} = -\frac{\sigma}{e} \nabla E_{ec}. \quad (21)$$

Using Eq. (6) one gets

$$\mathbf{j} = \sigma \mathbf{E} - D \nabla en' - \sigma \frac{e}{\kappa} \nabla \int \frac{n'(\mathbf{r}') d^3 r'}{|\mathbf{r} - \mathbf{r}'|}. \quad (22)$$

Here  $D$  is connected to  $\sigma$  by the Einstein relation [Eq. (4)]. Considering relaxation of the charge density one can ignore the external field  $\mathbf{E}$ . The relaxation is described by the continuity equation

$$\frac{\partial(en)}{\partial t} = -\nabla \cdot \mathbf{j} \quad (23)$$

or

$$\frac{\partial(en)}{\partial t} = \sigma \left( \frac{1}{e^2} \frac{d\mu}{dn} \nabla^2(en') - \frac{4\pi en'}{\kappa} \right). \quad (24)$$

The ratio  $R$  of the first (diffusion) term in the right-hand side to the second (field) term is  $R = (q_2^2 L^2)^{-1}$ , where

$$L = \sqrt{\left| \frac{n'}{\nabla^2 n'} \right|} \quad (25)$$

is the characteristic size of the extra charge and  $q_2^2$  is given by Eq. (11). If the electron gas is nonideal,  $q_3 \sim 1/\bar{r}$ , where  $\bar{r}$  is the average distance between electrons. However, the concept of diffusion equation is valid only at  $L \gg \bar{r}$ . This means that for the nonideal gas,  $|R| \ll 1$  and the diffusion term in Eq. (24) should be ignored. Then Eq. (24) has a simple solution,

$$n'(\mathbf{r}, t) = n'(\mathbf{r}, 0) \exp(-t/\tau_M), \quad (26)$$

where  $\tau_M = \kappa/(4\pi\sigma)$  is the well-known Maxwell time. The coefficient  $D$  does not enter in this case in the entropy production and the sign of  $D$  is not necessarily positive. Thus in a three-dimensional nonideal electron gas, negative  $d\mu/dn$  does not create any contradiction with the Einstein relation.

In the 3D gas of high density  $\mu \sim n^{2/3}$  and  $R \sim (\bar{r}/L)^2/r_s$  with  $r_s < 1$ . In this case  $R$  might be large and diffusion is possible. However  $d\mu/dn > 0$  and  $D > 0$ .

Now we consider the relaxation of the charge density in the two-dimensional case. Instead of Eq. (24) one gets

$$\frac{\partial(en_2)}{\partial t} = \sigma_2 \left( \frac{1}{e^2} \frac{d\mu}{dn_2} \nabla^2(en'_2) - \frac{e}{\bar{\kappa}} \nabla^2 \int \frac{n'_2(\mathbf{r}') d^2 r'}{|\mathbf{r} - \mathbf{r}'|} \right). \quad (27)$$

Here  $n_2$ ,  $\sigma_2$ , and  $\nabla$  are two-dimensional density, conductivity, and two-dimensional gradient, respectively. To discuss this equation it is convenient to make the Fourier transformation. Then one gets

$$\frac{\partial(n_q)}{\partial t} = -\sigma_2 \left( \frac{1}{e^2} \frac{d\mu}{dn_2} q^2 n_q + \frac{2\pi q}{\bar{\kappa}} n_q \right), \quad (28)$$

where  $n_q$  is the Fourier transform of  $n'_2$ .

Now we find that the ratio of the first (diffusion) term in the right-hand side of Eq. (28) to the second (field) term  $R_2 = q/q_2$ , where  $q_2$  is given by Eq. (13). Similar to the 3D case, in the nonideal gas  $|q_2| \sim 1/\bar{r}$  and diffusion should be ignored. Then we get the Dyakonov-Furman equation,<sup>20</sup>

$$\frac{\partial(n_q)}{\partial t} = -v q n_q, \quad (29)$$

where velocity  $v = 2\pi\sigma_2/\bar{\kappa}$ . The physical meaning of this equation is that the extra density of electrons localized initially at some spot propagates in all directions with velocity  $v$  conserving the total amount of extra electrons. Of course, this way of relaxation is more efficient than diffusion (random walk) because  $r \sim vt$  while  $r \sim \sqrt{Dt}$  in the case of diffusion. Thus, the diffusion coefficient  $D$  is irrelevant and nega-

tive  $d\mu/dn$  does not create any contradiction with the Einstein relation. In a high-density electron gas,  $R_2=q\bar{r}/r_s$  and the diffusion mechanism is possible. In this case  $d\mu/dn > 0$  and  $D > 0$ .

One can consider this problem from a different point of view. In both 3D and 2D cases the negative diffusion coefficient  $D$  appears in the term with the highest derivative that leads to the absolute instability even if  $D$  is small.<sup>21</sup> Consider, for example, Eq. (24), the 3D case. After the Fourier transformation the solution for the charge density  $\rho=en'$  can be written in the form

$$\rho_q = \rho_q^0 \exp\left(-\frac{4\pi\sigma t}{\kappa} - Dq^2 t\right), \quad (30)$$

where  $D$  is given by the Einstein relation [Eq. (4)]. One can see that at  $D < 0$  solution increases with time exponentially for harmonics with  $q\bar{r} \geq 1$ .

The physical explanation is as follows. Equations (24) and (27) contain the average distance between electrons  $\bar{r}$ . So they contain information that the charged liquid has a discreet electronic structure. This information comes from the negative density of states which originates from the interaction of the separate electrons. That is why macroscopic equations become unstable at large  $q$ . The message is that  $n(\mathbf{r})$  is rather a set of  $\delta$  functions than a continuous function. The instability is absent if  $D$  is positive.

The instability at large  $q$  and small negative  $D$  does not affect harmonics with small  $q$  because Eqs. (24) and (27) are linear. Due to the linearity different harmonics are independent and any exchange of energy between them is forbidden (cf. phenomenon of turbulence in nonlinear hydrodynamics where the exchange of energy is not forbidden, but the instability is initiated at large distances). Therefore, we conclude that at small  $D$ , the approximation  $D=0$  that gives Eqs. (26) and (29) is correct.

One should note that the problem of the nonphysical roots of electric permittivity discussed in Sec. II is of the same nature. Before, we have discussed unipolar diffusion. Consider now the simplest case of ambipolar diffusion assuming that at  $t=0$  the densities of electrons and holes are equal in some finite region of space and are zero otherwise. Moreover we assume that the local macroscopic charge density  $\rho(\mathbf{r}, t) = 0$  and the recombination of carriers is very slow. In this case Eq. (6) describes the electron-hole system in quasiequilibrium. At large  $r_s$  one gets  $E, \mu, d\mu/dn < 0$  but the last term in Eq. (6) is absent. So the smearing of the density of particles is described by the equation of diffusion at all  $r_s$ , but at small density ( $r_s \geq 1$ ), the coefficient  $D < 0$ . Then the absolute instability takes place for all harmonics that means a collapse of the system. Thus the electron-hole ‘‘Wigner liquid’’ and crystal are unstable.

This result is very transparent. It happens because negative  $\mu$  just means that the energy of the system decreases with increasing density. In the bipolar case, neutrality is provided by the particles and we do not consider any background. Thus the instability is a result of the negative compressibility in a neutral system. At large enough  $r_s$  these particles are classical, and the absence of the mechanical

equilibrium follows also from the Earnshaw theorem. In reality quantum mechanics becomes more important with increasing density. As a result excitons are formed. These neutral particles have a positive diffusion coefficient  $D_a$  and their density smears with time through all available space. This process is described by a regular diffusion equation. In the case of optical excitation the carriers may appear in the form of the excitons from the very beginning

For the coefficient of ambipolar diffusion  $D_a$  a textbook equation<sup>22</sup>

$$D_a = \frac{2D_e D_h}{D_e + D_h} \quad (31)$$

is often used, where  $D_{e,h}$  are the unipolar diffusion coefficients of electrons and holes. As follows from the previous discussion, one should be careful with this equation because for the nonideal electron (or hole) gas these unipolar coefficients might be negative and meaningless. That happens because in the unipolar case there is a deviation from neutrality that creates an electric field, while in bipolar case the system is neutral. In this case Eq. (31) does not work and one should calculate  $D_a$  in a different way as a diffusion of the excitons.

In the recent paper by Zhao<sup>23</sup> the experimental results for ambipolar diffusion in a silicon-on-insulator system are compared with Eq. (31). At high temperatures a good agreement is found, while at low temperatures the observed values of  $D_a$  are six to seven times less. The previously reported values<sup>24</sup> show similar temperature dependence.

The author’s explanation is that coefficients  $D_{e,h}$  are taken for the bulk silicon using Einstein relation and they might be larger than in the film at low temperatures. However, the reason discussed above cannot be excluded.

#### IV. GRAPHENE AS A POSSIBLE EXAMPLE OF A NONIDEAL ELECTRON SYSTEM

It is interesting to discuss the single layer graphene as an example of the system with nonideal electron gas. Graphene is a gapless material with a linear spectrum of electrons and holes near the Dirac point. Due to some reasons, which are not quite clear now, the velocity  $v$  of electrons and holes in equation  $\epsilon = \pm pv$  is of the order of  $e^2/\hbar$ . It follows that at any Fermi energy within this linear spectrum, the electron gas in graphene is nonideal in a sense mentioned above: the absolute value of the chemical potential is of the order of interaction energy  $e^2 n^{1/2}$ . This means that unipolar density relaxation in this system should be described by the Dyakonov-Furman equation<sup>20</sup> rather than by the diffusion equation.

However, without magnetic field the electron gas in graphene is marginally nonideal. It cannot be classical, like an electron gas of low density with a quadratic spectrum. The marginal situation makes theoretical calculations very difficult. Nevertheless, it is accepted that the Wigner crystal in single layer graphene is absent without magnetic field.<sup>25</sup> The sign of  $d\mu/dn$  is also an interesting question but very difficult for theoretical study. Recently tunneling microscopy experiment has been done by Martin *et al.*<sup>26</sup> They claimed that their measurement gives the thermodynamic density of



states and that it is positive. The last statement might be a result of disorder.

## V. CONCLUSION

Finally I argue that the negative sign of diffusion coefficient that follows from the Einstein relation at negative density of states does not lead to any contradiction because the diffusion coefficient is irrelevant for unipolar transport under this condition. The sign of the diffusion coefficient in this

case should not be definitely positive because the diffusion is not the main source of the entropy production. In the bipolar situation negative diffusion means the collapse of the system and formation of neutral excitons.

## ACKNOWLEDGMENTS

I am grateful to Boris Shklovskii, Serge Luryi, and Yoseph Imry for important discussion. I am especially indebted to David Khmel'nitskii and Emmanuel Rashba for multiple discussions and criticism.

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