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1990 J. Phys.: Condens. Matter 2 9055

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Frequency-dependent conductivity in glasses

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Received 8 May 1990, in final form 16 July 1990

Abstract. The Barton–Nakajima–Namikawa relationship between the static dielectric constant, $\varepsilon(0)$, and the DC conductivity $\sigma(0)$, observed in many glasses, $\sigma(0) \propto \omega_c \varepsilon(0)$, with ω_c the loss peak frequency, has been generally assumed to signify a connection between the low frequency AC and the DC conduction processes. We argue that the connection is that both are due to non-local relaxation processes, the relaxation time of the DC process being effectively infinite. Above the loss peak, the relation $\sigma(\omega) \propto \omega^s$ is compatible with the pair approximation, i.e. a local relaxation theory. Below the loss peak, the role of Coulomb interactions is critical. If the effects of Coulomb interactions may be neglected, or treated perturbatively, the relaxation time of a process spanning a linear dimension x is proportional to x^2 . But if Coulomb interactions dominate, the relaxation time may be proportional to x. Only the latter condition is compatible with the BNN relation, and is believed to be one of the most important distinctions between the ionic glasses and the Fermi (electronic) glass.

1. Introduction

In treating conduction processes of ionic glasses, a clear distinction has usually been drawn between polarization processes (frequency-dependent conductivities and dielectric constants) and steady state transport processes (DC conductivity). The former are often treated in either a Debye or more complex relaxation theory, the latter by calculating a typical structural activation energy. It has often been argued that a distribution of relaxation times is necessary to explain the higher frequency behaviour (the pair approximation regime, Pollak and Geballe 1961, Austin and Mott 1969, Pollak and Pike 1972). It has recently been established (Hunt and Pollak 1990, Hunt 1990a, b) that relaxation processes of all posible relaxation times, from inverse phonon frequencies to infinity play a role in the frequency-dependent transport in the electronic system in amorphous semiconductors ('Fermi glass'), and as well in the Coulomb gap in impurity conduction systems (the 'electron glass'). We argue here that a treatment of ionic transport incorporating this feature is probably necessary. As a result, the low frequency relaxation is seen to have non-local characteristics, and a conceptual link is established between DC and low frequency AC processes. In fact, the DC conductivity may be regarded as arising from processes with infinite relaxation times. Thus an arbitrary upper limit on relaxation times is unjustifiable. Moreover, the role of Coulomb interactions in the relaxation process is emphasized, as argued by Jonscher (1977, 1980, 1981), Hill and Jonscher (1983) and Ngai (1984). At high frequencies we expect that a pair approximation approach is indeed appropriate. The main criticism of the pair approximation,

as given by Dyre (1986, 1988) and Jonscher (1977), is that the exponent s in $\sigma(\omega) \propto \omega^s$ appears to increase at higher frequencies, if it varies at all, in contradiction to the prediction that s should decrease. This criticism, however, has been made by confusing the near DC region on the log-log plot of $\sigma(\omega)$ with the lower portion of the pair approximation regime, at least in the case of a-Si. (See also the conclusions.) This region is not described by the pair approximation, because to apply the pair approximation pairs with rates $w \approx \omega$ must be able to respond much more rapidly to the applied field than the remainder of the system. In many cases it is evident that the loss peak defines the lower limit of applicability of the pair approximation (in the electronic glasses this has been shown explicitly, see the references to Hunt); it therefore represents the onset of the importance of non-local relaxation processes. This point of view is not entirely new, to the extent that the behaviour of $\sigma(\omega)$ above the loss peak has been shown to be independent of the behaviour below the loss peak (Jonscher 1980). What is new is the implication that non-local relaxation processes are dominant below the loss peak, and that a pair approximation should suffice above the loss peak. In the non-local relaxation processes, in particular, in ionic conducting glasses, Coulomb interactions are expected to play an important role.

The model presented here is sufficient to extract the Barton (1966)–Nakajima (1971)– Namikawa (1975) (BNN) relation, the position asymmetry, and typically large width of the loss peak, as well as approximate frequency dependences for $\sigma(\omega)$ above and below the loss peak. It is consistent, as well, with an activated DC conductivity. It must be emphasized, however, that this treatment is still somewhat superficial in its application to ionic conduction in glasses; as a consequence areas for improvement in the theoretical treatment are pointed out.

2. Model

We assume, similarly as in Dyre (1986, 1988), an exponential dependence of individual relaxation times τ_i on energy barrier heights,

$$\tau_i = \tau_0 \exp(-E_i/kT) \tag{2.1}$$

and assume also that E_i is a random variable. Here τ_0^{-1} is on the order of a phonon frequency, roughly $10^{12}-10^{13}$ Hz. The width of the distribution of E values is roughly equal to kT', where T' is the temperature at which $\sigma(\omega)$ becomes frequency-independent up to frequencies nearly on the order of phonon frequencies. For $kT \ll kT'$ the large spread in individual relaxation times allows an application of percolation theory to the calculation of the DC conductivity. As the only random variable is E, and not the length of the hop, r (as activation over a barrier requires no tunnelling factor), the percolation theory could consistently be called E-percolation. This process will yield an activated conductivity, as a limiting process with a specific barrier height will define the critical rate for DC transport. Provided the distribution of barrier heights is independent of temperature, this activation energy will also be temperature independent.

At relatively large frequencies, the pair approximation (Pollak and Geballe 1961, Hunt 1990c) is quite generally based on the assumption that at frequency ω processes with $\tau = 1/\omega$ dominate the conductivity, and that individual pair processes may be treated independently from one another. Such a treatment also requires a wide range of individual relaxation times; a fact which is compatible with the condition for the application of percolation theory to the DC conductivity. In glasses the starting point for the pair approximation is the following equation,

$$I_{\text{pair}} \propto \tau_{\text{pair}}^{-1} (\omega^2 \tau_{\text{pair}}^2 + i\omega \tau_{\text{pair}}) / (1 + \omega^2 \tau_{\text{pair}}^2)$$
(2.2)

in which the AC current flowing in a pair, I_{pair} , is proportional to the product of an inverse of the relaxation time of the pair, τ_{pair} , and a frequency dependent factor appropriate for the conductance of a resistor and capacitor in series. This current is maximized when $\tau \propto 1/\omega$; treating only isolated processes satisfying this condition yields $I(\omega) \propto \omega$, if the distribution of barrier heights is independent of barrier height. Otherwise the actual logarithmic dependence of the random variable E on τ introduces logarithmic corrections in the dependence on the frequency ω , making the functional dependence slightly sublinear at frequencies well below τ_0^{-1} . In analogy with the electronic glasses, we expect a frequency dependent conductivity at high frequencies that is approximately given by the pair approximation. As ω is reduced, the density of pairs for which $\tau < 1/2$ ω increases, until at some point the approximation that the relaxation processes may be treated independently breaks down. As has been demonstrated by Hunt (1990c), in the Fermi glass it is possible to formulate a pair approximation which breaks down at a frequency proportional to the DC conductivity. At this frequency, the pair separation is approximately equal to the pair length. Also at this frequency, which is evidently approximately equal to the loss peak frequency, the pair and the low frequency approximations yield the same temperature dependence of the conductivity. In ionic glasses, we assume that a similar principle is in operation; in particular, at frequencies $\omega \propto \sigma_{DC}$, the separation of pairs with $\tau < 1/\omega$ is on the order of the hopping lengths. This assumption is compatible with the assumption that a critical rate, τ^{-1} , exists which defines a DC conducting 'network'. This viewpoint is compatible with the literature, as Butcher and Morys (1973), for example, have noted that the pair approximation is exact in the limit of low pair density.

A rather frequently used procedure has been to include the entire system in a meanfield approximation. In the Fermi glass, the system is representable by the Miller-Abrahams (1961)-Pollak (1974) random impedance network. In the extended pair treatment of this network (Summerfield and Butcher 1982), for example, an arbitrary pair of sites is embeddeded in a medium of complex admittance Y, and the effective admittance Y_{eff} of the pair plus the medium is averaged over all possible pairs under the self-consistency condition, $Y_{eff} = Y$. This method generates an asymmetric loss peak, the appropriate DC conductivity, the BNN relation, the high frequency pair result, and an analytic, i.e. quadratic frequency dependence of the real part of the conductivity, $\text{Re } \sigma(\omega)$, below the loss peak. Since, however, the frequency dependence of $\sigma(\omega)$ below the loss peak in the electronic glass is not quadratic (as it isn't in virtually all ionic glasses, and many organic systems as well), and since, moreover, the BNN relation has not been established in the Fermi glass (Long *et al* 1988), it seemed preferable to avoid the approximations inherent in a mean-field approach, which moreover also neglects the dynamic effects of Coulomb interactions.

Although the model for the low frequency conduction process in glasses is still sketchy, many parallels to glassy electronic systems exist and can be exploited. The electronic systems can be classified according to whether or not the inter-site Coulomb interactions dominate. The former system is termed the electron glass, the latter, the Fermi glass. The importance of Coulomb interactions can be determined by comparing the Coulomb gap width (in the single electron density of states) with kT. At very low temperatures the opening of this gap in the single-electron density of states corresponds

to the increasing importance of Coulomb interactions. At higher temperatures, the glassy nature of the electronic system is due to the presence of very high disorder. The low frequency conductivity of the Fermi glass is strongly affected by the large scale inhomogeneities in the impedance network. A powerful advantage of the random impedance network is that the statistics of clusters of impedances are given in terms of percolation theory (Stauffer 1990).

At lower temperatures, the Coulomb interaction is known to make the electronic system virtually spatially homogeneous in the vicinity of the Fermi energy, $E_{\rm F}$. In this case, the low frequency conductivity is very strongly affected by sequential correlations of individual transitions, also arising from Coulomb interactions. This latter situation is more closely analogous to the situation in ionic glasses, but a rigorous correspondence between electronic transport and a well-defined impedance network has only been established for the so-called 'Fermi glass'. Thus the subsequent discussion will utilize analogies from both systems, but it must be admitted that no concrete definition of resistances and capacitances for the ionic glasses is available.

An interesting contrast between the 'electron glass' and the 'Fermi glass' is that the DC current in the Fermi glass (Hunt 1990a) is carried by the DC cluster; large scale inhomogeneities can be represented by large regions of space with a higher conductivity (at frequency ω). The electron glass, on the other hand has no rigidly defined clusters. A calculation of the time dependent current demonstrates that the polarization current derives from the temporal development of the clusters (Hunt 1990b). At the same time this constant growth of the clusters in which equilibrium has been reached supports a steady state current in the clusters as well. Thus there is no distinction between steady state (DC) clusters and polarization (AC) clusters. This point of view can be shown to be consistent with the assumption (made later) that the static polarizability of a cluster of N elements, as well as the effective resistance, do not depend on N. It also allows the representation of the total AC conductivity, $\sigma(\omega)$, as $\sigma(\omega) = \sigma_p(\omega) + \sigma_{DC}$, where the polarization current density calculated here defines the non steady-state contribution, $\sigma_p(\omega)$, to the total conductivity.

In the above described new theory the frequency dependent polarization currents in clusters of impedances have been calculated and summed over all clusters to get the frequency dependent conductivity. This theory has been much more successful in explaining the low frequency dependence of $\sigma(\omega)$ and $\varepsilon(\omega)$ on the frequency and on the temperature. Moreover, it yields known theoretical results in one-dimensional systems. It only generates the BNN relation in systems where the effect of large scale inhomogeneities is severely modified through the effects of Coulomb interactions or for d > 6, the upper critical dimension for percolation theory. We believe that these conditions indicate its usefulness for describing conduction in ionic glasses, but it points up a problem in the choice of a specific form for the statistical distribution of large clusters. In the electronic glasses percolation statistics are used, but some modification of these statistics may be required in the Coulomb interaction correlated systems. These questions will become clearer as the discussions develops.

When relaxation processes considered are non-local, it is necessary first to determine how much charge can be transported by the dominant processes with relaxation time τ , and how far the charge is transported. The condition that relaxation processes with $\tau \approx 1/\omega$ dominate holds for low frequencies just as it does for the pair approximation regime. In the electronic system discussed above the problem is well-defined. It has been shown that a cluster of similar maximally valued impedances R, with individual relaxation times, $\tau = RC = Re^2/kT$, where C is a typical capacitance per maximally valued resistance, has a maximum relaxation time $\tau_N = N^2 \tau$. Here N is the number of maximally valued resistances R on a one-dimensional path spanning the cluster (Hunt and Pollak 1990). The enhancement factor N^2 can be understood as the product of the number of charges transported through the chain (proportional to N) and the number of rate limiting links (also proportional to N). This result has been demonstrated to hold also when Coulomb interactions in a screening approximation are taken into account. It has also been shown (Pollak and Pohl 1975) that the current flowing in such a chain (to within 20% accuracy in the worst case) can be represented by the single (slowest) relaxation process,

$$I_N(\omega) = F_0(Nl)^2(NR)^{-1}(\omega^2 \tau_N^2 + i\omega \tau_N)/(1 + \omega^2 \tau_N^2)$$
(2.3)

with the external field $F = F_0 \cos \omega t$, $(Nl)^2$ the static polarizability of the (one-dimensional) path, and NR its effective resistance. To a good approximation (Hunt 1990a) it is then possible (except in one dimension) to set

$$\sigma(\omega) = \sigma_{\rm DC} + \sigma_{\rm p}(\omega) \tag{2.4}$$

where $\sigma_{\rm DC}$ is proportional to the inverse of the critical (percolation) resistance, $R_{\rm c}$, and $\sigma_{\rm p}(\omega)$ is

$$\sigma_{\rm p}(\omega) = F_0^{-1} \sum I_{N,R}(\omega) n_{N,R} \tag{2.5}$$

with $n_{N,R}$ defining the number of one-dimensional paths in which N maximally valued resistors of magnitude R are connected in series. If the statistics of $n_{N,R}$ are given in terms of the critical resistance, as in percolation theory, $\sigma_p(\omega)$ is automatically related to σ_{DC} .

In the electron glass, the proportionality of τ_N to N^2 does not hold. A treatment of the additional contribution to the free energy arising from the application of a very low frequency field has shown that the relation $\tau_N \propto N$ is appropriate. This result is due to the ordering of the electrons brought about by the strong Coulomb interactions. We believe that the same result is likely to hold here. We assume (as is usual) that ionic hopping distances are usually only one or two lattice constants (i.e. on the order of or less than the ionic separation), and (as is also usual) that interactions are very important. In particular the concentration of ions may be 10^4 larger than the density of electrons in gap states in, e.g., a-Si and a-Ge, so that typical electric field strengths due to interactions may be enhanced by a factor $(10^4)^{2/3} \approx 600$. Thus the range of temperatures for which the effects of Coulomb interactions are crucial is much larger than in the electronic systems, where effects due to the Coulomb gap are observed at temperatures below a few Kelvin. For this reason we think it probable that ionic glasses the hops of individual ions are strongly sequentially correlated over a much wider temperature range, in fact over the same temperature range that the high frequency AC conductivity is given by the pair approximation. In this picture the number of ions passed by the rate limiting links of a cluster is more nearly independent of the length of the cluster (in contrast to the non-interacting case, where the proportionality was to N^2). A perfect sequential correlation of the hops would nevertheless result in a relaxation time proportional to N. Corresponding to the reduction in the relaxation time as compared to the non-interacting system, we expect to find a reduction in the effective resistance by a factor N, and in the static polarizability by a factor N^2 since each rate limiting process is traversed by a number of charges independent of N. As a consequence the static dielectric constant is reduced by a factor N^2 , guaranteeing its convergence (except in one dimension) in contrast to the Fermi glass. We now write for the current in such a cluster (keeping in mind that the quantity R is not well-defined in this context)

$$I_{N,R} = (l^2/R)(\omega^2 \tau^2 N^2 + i\omega\tau N)/(1 + \omega^2 \tau^2 N^2)$$
(2.6)

in analogy with (2.3) for the Fermi glass. Here l is the typical separation of the slowest processes. If some variation of percolation statistics is used, in which the cluster sizes are defined in terms of a critical resistance and the typical separation of these resistances, the product of the conductance R^{-1} of this bond and the appropriate inverse length (i.e. the separation l) will yield roughly the (activated) DC conductivity.

In contrast to the Fermi glass, in which the chain current $I_{N,R}(\omega)$ is a strongly increasing function of N, the result (2.6) has a prefactor independent of N. Therefore the approximation made in the Fermi glass, that each cluster could be replaced by its longest chain, does not apply here. To find $n_{N,R}$, then we take a sum over the product of the distribution of clusters with m as the longest chain length, and the distribution of chain lengths N on a given cluster with m as a maximum chain length. It can be shown (Hunt 1990) that the number of all N sequences with individual τ values equal to the critical (DC) value is

$$n_N \approx l^{-3} N^{-2-d+d_{\rm f}} \tag{2.7}$$

with *l* a typical separation of the rate limiting processes, and where d_f is the fractal dimensionality associated with percolation in *d* dimensions. Note that in the quoted reference the roles of the symbols *m* and *N* are reversed. In one dimension this equation (2.7) yields $n_N \propto N^{-2}$, since $d = d_f = 1$, which agrees with known results. In higher dimensions, the difference between *d* and d_f is small ($d_f = 2.5 \text{ in 3D}$, 1.9 in 2D), so that n_N does not vary strongly with dimension. We emphasize that this approach may be incorrect, as the result was derived from the assumption of the self-similarity of the percolation path. On the other hand, it is clear that the result $n_N \propto N^{-2}$ implies and is implied by one-dimensional transport. It is also clear that the density of large clusters must be reduced over the one-dimensional value if the transport is through clusters which have two or three dimensional structure. In fact, however, this is one of the critical points at which a more in depth analysis may result in modifications of the theory.

It is interesting that any distribution of n_N whose first moment converges will preserve the BNN relation, although different powers of ω result. But note again the exception in one dimension for which $n_N \propto N^{-2}$. In this case, as we will see, the dielectric constant will diverge logarithmically in the limit of zero frequency and the BNN relation will not be obtained.

Proceeding, we have,

$$\sigma_{\rm p}(\omega) = (lR)^{-1} \sum N^{d_{\rm f}-d-2} (\omega^2 \tau_{\rm c}^2 N^2 + i\omega \tau_{\rm c} N) / (1 + \omega^2 \tau_{\rm c}^2 N^2)$$
$$= \sigma_{\rm DC} \int dN N^{-2-d+d_{\rm f}} (\omega^2 \tau_{\rm c}^2 N^2 + i\omega \tau N) / (1 + \omega^2 \tau_{\rm c}^2 N^2).$$
(2.8)

Here the symbol τ_c represents a critical relaxation time, for which it is possible to just find a macroscopic cluster of relaxation times $\tau < \tau_c$. Use of this value for τ is justified by the application of percolation cluster statistics for clusters with the critical rate. The result for the AC conductivity is

$$\sigma(\omega) = \sigma_{\rm DC} + \sigma_{\rm DC} [(\omega/\omega_{\rm c})^{1+\delta} + i(\omega/\omega_{\rm c}) - i(\omega/\omega_{\rm c})^{1+\delta}]$$
(2.9)

with $\delta = d - d_f$ the anomalous dimensionality, and where numerical factors have not

been included, but are of order one in the leading terms. Obviously the numerical factor of the second term in the imaginary part of the conductivity, Im $\sigma(\omega)$ is considerably smaller than in the first term. The result for the dielectric constant, $\varepsilon(\omega)$ is

$$\varepsilon(\omega) = (\sigma(\omega) - \sigma_{\rm DC})/i\omega = \sigma_{\rm DC}/\omega_{\rm c}\{[1 - (\omega/\omega_{\rm c})^{\delta}] - i(\omega/\omega_{\rm c})^{\delta}\}$$
(2.10)

leading to (as long as d > 0)

$$\operatorname{Re}\varepsilon(0) \propto \sigma_{\rm DC}/\omega_{\rm c} \tag{2.11}$$

for the static dielectric constant, $\varepsilon(0)$, which is essentially the BNN relation. An equivalent statement is that the loss peak frequency scales with temperature exactly as does the DC conductivity. This statement can be understood if one considers the following. Results for $\sigma(\omega)$ for frequencies higher than the loss peak frequency generated by application of the pair approximation become invalid near the critical frequency, ω_c (because of the formation of clusters of processes of relaxation times τ_c), and yield at this frequency approximately the same absolute value of the conductivity as does the low frequency result. As a consequence, the critical frequency may be approximately identified with the loss peak frequency.

In case transport is along purely one-dimensional paths, the low frequency conductivity is exactly linear in the frequency (as in the electron glass), and the dielectric constant acquires a logarithmic divergence in the limit of zero frequency.

Note that the sublinear frequency dependence in Im $\varepsilon(\omega)$ can account for a broad loss peak. Moreover, the power on the low frequency side of the loss peak, d, is unrelated to the power on the high frequency branch, s - 1, typically a small negative number, since $\sigma(\omega)$ in the pair approximation is slightly sublinear. Thus asymmetry is a general property of such a treatment as the processes on both sides of the loss peak are distinct.

We leave the expression for $\sigma(\omega)$ in the above rather general form. It is suggestive that the dimensionality of the conducting paths may determine the exponent on the conductivity. In this vein, we mention that Namikawa's study of the so-called BNN relationship found that virtually all glasses satisfied $\sigma_{DC} = B\omega_c \varepsilon(0)$, but with two different values of *B*. The value of *B* obtained from the above treatment is dependent on *d*, and this correspondence is suggestive. Furthermore, powers of the frequency compatible with those derived have been observed. It is also suggestive that a third class of ionic glasses exist for which the BNN relationship is not satisfied. In particular, a roughly logarithmic divergence in the dielectric constant has been measured even to the lowest accessible frequencies, although, in fact a kink in $\varepsilon(\omega)$ is seen at frequencies corresponding to the loss peak frequency.

Nevertheless, a correspondence between this theory and experiment should not be regarded as conclusively established. Both theoretical and experimental uncertainties exist. In the first place, although the values of B seem to fall into two classes, no corresponding universality in the powers of the frequency in the frequency dependent dielectric constants and the conductivity has been noted. Furthermore, the universality implied by percolation statistics has been derived specifically for clusters of impedances which are not strongly correlated by Coulomb interactions. This degree of universality may not be appropriate for ionic glasses, although one might argue that an even higher degree of universality would be expected. Finally, the assumption that the relaxation time of a cluster of individual rate limiting process should depend linearly on the number of such processes connected in succession, is an assumption. Although it can be

supported more rigorously in the case of the electron glass, for which a very specific model exists, it has not been verified here.

3. Conclusions

We have shown that the objections to the application of the pair approximation to the relatively high frequency conductivity of glasses can be removed, or are in error. These objections have been: (i) that the power s in ω^{s} increases with frequency, if it changes at all, (ii) that the pair approximation implies a zero DC conductivity, (iii) that the loss peak is not a feature of the pair approximation, d, that the pair approximation does not reproduce the BNN relation. The last three objections can be removed if the pair approximation is applied only in the region for which it is valid, namely when the density of pairs treated is small, but is replaced by a suitable modification when this condition is violated (as in this work). Objection (i) can be seen to be false if one examines closely the data quoted by Jonscher. In particular, we consider figure 2 in the Nature article (1977) which has been reproduced elsewhere, and referred to by Dyre as well. Jonscher claims that the power of s in three cases increases with ω , specifically a-Si, β -alumina, and stearic acid. The first case has been investigated much more carefully by Long (1988). In fact the region to which Jonscher refers is near the DC value of $\sigma(\omega)$, and the rounding on the log-log scale represents chiefly the constant term in $\sigma(\omega)$, namely $\sigma_{\rm DC}$. This frequency range actually exhibits (Long 1988) a larger power than at higher frequencies and corresponds to the lower branch of the loss peak when the data for the dielectric constant are analyzed. For this range of frequencies the pair approximation is inadequate. In fact the treatment referred to here is appropriate for this range of frequencies, and the agreement is found to be superior to that of other theories, such as the extended pair approximation of Summerfield and Butcher (1982) and Summerfield (1985). Nevertheless, the extended pair approximation is in agreement with this theory in the rough generalities.

We have shown that application to the AC conductivity of a theory incorporating non-local relaxation effects is consistent with the general trends of experiment in a wide variety of systems. It reproduces the BNN relation in ionic glasses where the role of Coulomb interactions is known to be extremely important. It also gives results for $\sigma(\omega)$ which are compatible with experiment in specific electronic conduction systems, in particular in the 'Fermi glass' and in the 'electron glass'. It has been shown that under conditions where the Coulomb interactions may be neglected, or treated perturbatively, the BNN relationship does not result. When Coulomb interactions are strong enough to sequentially correlate individual transitions, the BNN relationship obtains, unless the transport is restricted to purely one-dimensional channels. As a consequence the role of Coulomb interactions in the derivation of the BNN relationship is emphasized

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