Importance of exchange contributions to scattering and screening in the quantum kinetic regime

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Abstract. We study the role of exchange contributions for the ultrafast carrier relaxation in the quantum kinetic regime. The analysis is based on a density matrix approach where besides the direct contributions to Coulomb scattering and screening in the random phase approximation (RPA) also the corresponding exchange terms are included which are necessary to satisfy the correct antisymmetry of the two-particle density matrix. At early times we find significant deviations from the commonly used RPA where these exchange contributions are neglected. At later times exchange contributions to scattering and screening tend to compensate each other and the differences between RPA and the calculations including exchange are reduced.

1. Introduction

The quantum kinetics of a gas of charged particles on ultrashort time scales has been a very active research field in the past years first because the description of modern optical experiments using pulses in the range of a few to a few tens of femtoseconds requires a detailed modeling of the carrier dynamics and second because here fundamental phenomena like memory effects, the energy-time uncertainty or the build-up of screening can be studied. Indeed it has been found that at very early times the dynamics is well described by scattering processes via an unscreened potential [1, 2] while with increasing time the screening is built up [3, 4, 5] resulting in a reduced scattering efficiency. This build-up of the screening has been studied in sophisticated quantum kinetic calculations based on fully time-dependent RPA dielectric functions [4] and good agreement with experiments in the ultrafast regime has been obtained [6, 7]. In these calculations, however, which are mostly based on a nonequlibrium Green's function approach exchange effects have been neglected because they correspond to a different class of diagrams beyond RPA.

An alternative approach to quantum kinetic phenomena on ultrashort time scales is the density matrix theory which has been extensively applied mainly to electron-phonon quantum kinetics as well as to the Coulomb quantum kinetics in the low-density regime. It is well known already since the early 1960s that also here, if two-particle correlations are included, the full dynamical RPA screening of the scattering rates is obtained [8, 9, 10]. In recent years several authors have addressed the treatment of screening in this formalism [11, 12, 13, 14], however it has scarcely been applied to numerical investigations. Interestingly, in this approach the exchange contributions appear in a natural way on the same level as the direct contributions. In fact, it is obvious from the equations that they are necessary to obtain two-particle density matrices which fulfill the correct Fermi antisymmetry. Exchange terms modify both the scattering and the screening contributions to the dynamics of two-particle correlations and it is the interplay between these two phenomena which is the focus of our investigations. Exchange contributions to the carrier relaxation have been studied in terms of

semiclassical scattering rates [15, 16, 17]. This approach, however, does not hold at ultrashort times where quantum kinetic effects are important.

In this contribution we study the role of the quantum mechanical exchange for the ultrafast relaxation of a two-dimensional electron gas due to Coulomb interaction processes based on the density matrix theory. By selectively switching on and off the contributions corresponding to RPA screening, exchange scattering, and exchange screening we can clearly identify the role of these contributions for the relaxation dynamics. To concentrate on the relaxation process we limit ourselves to a one-band model for a quasi-two-dimensional electron gas where we analyze the dynamics of a given initial distribution.

2. Theory

We consider the standard model for a two-dimensional interacting electron gas. The material parameters are taken for the conduction band of a GaAs quantum well. For this system the Hamiltonian is given by

$$H = \sum_{\mathbf{k},\sigma} \varepsilon_{\mathbf{k}} c^+_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \frac{1}{2} \sum_{\mathbf{k},\mathbf{k}',\mathbf{q},\sigma,\sigma'} V_{\mathbf{q}}\, c^+_{\mathbf{k}\sigma} c^+_{\mathbf{k}'\sigma'} c_{\mathbf{k}'+\mathbf{q}\sigma'} c_{\mathbf{k}-\mathbf{q}\sigma}. \tag{1}$$

Here, $c^+_{\mathbf{k}\sigma}$ and $c_{\mathbf{k}\sigma}$ denote creation and annihilation operators for an electron with in-plane momentum $\mathbf{k}$ and spin index σ, $\varepsilon_{\mathbf{k}}$ is the free carrier dispersion relation assumed to be parabolic and $V_{\mathbf{q}}$ is the Coulomb matrix element screened only by the background dielectric constant. In a density matrix approach Coulomb scattering and screening is described by a coupled set of equations for the one-particle density matrix (single particle distribution function) $f^{\sigma}_{\mathbf{k}}$ and the two-particle density matrix $K^{\sigma\sigma'\sigma'\sigma}_{\mathbf{k}\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}\,\mathbf{k}-\mathbf{q}}$ defined as

$$f^{\sigma}_{\mathbf{k}} := \langle c^+_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} \rangle, \tag{2}$$

$$K^{\sigma\sigma'\sigma'\sigma}_{\mathbf{k}\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}\,\mathbf{k}-\mathbf{q}} := \langle c^+_{\mathbf{k}\sigma} c^+_{\mathbf{k}'\sigma'} c_{\mathbf{k}'+\mathbf{q}\sigma'} c_{\mathbf{k}-\mathbf{q}\sigma} \rangle, \tag{3}$$

respectively. In the spirit of the correlation expansion it is useful to define a correlation function $\bar{K}$ by subtracting from the two-particle density matrix K its mean-field part. Here we concentrate on spin-independent distributions $f_{\mathbf{k}}$ as generated, e.g., by unpolarized or linearly polarized light pulses which allows us to define

$$\bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} := \sum_{\sigma'} K^{\sigma\sigma'\sigma'\sigma}_{\mathbf{k}\,\mathbf{k}'\,\mathbf{k}'+\mathbf{q}\,\mathbf{k}-\mathbf{q}} + f_{\mathbf{k}} f_{\mathbf{k}-\mathbf{q}} \delta_{\mathbf{k},\mathbf{k}'+\mathbf{q}}. \tag{4}$$

As is typical for a many-body problem, by using the Heisenberg equations of motion for the creation and annihilation operators an infinite hierarchy of equations for higher order density matrices is obtained which, in the present case, represents the quantum BBGKY hierarchy. A common approximation to truncate this hierarchy is to neglect any three-particle correlations which results in a closed set of equations for $f_{\mathbf{k}}$ and $\bar{K}$ [10, 11, 12, 13, 14]. The remaining terms can be interpreted on physical grounds [10]. By keeping all terms relevant for a fully dynamical treatment of scattering and screening and satisfying at the same time the correct antisymmetry of the two-particle density matrix, the resulting set of equations of motion reads

$$i\hbar \frac{d}{dt} f_{\mathbf{k}} = 2i \sum_{\mathbf{k}',\mathbf{q}} V_{\mathbf{q}} \,\mathrm{Im}\left\{ \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} \right\}, \tag{5}$$

$$i\hbar \frac{d}{dt} \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} = \left(\mathscr{E}_{\mathbf{k}-\mathbf{q}} + \mathscr{E}_{\mathbf{k}'+\mathbf{q}} - \mathscr{E}_{\mathbf{k}'} - \mathscr{E}_{\mathbf{k}} \right) \bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}} \tag{6}$$

$$+ 2V_{\mathbf{q}} \left[f_{\mathbf{k}} f_{\mathbf{k}'} \left(1 - f_{\mathbf{k}'+\mathbf{q}} \right) \left(1 - f_{\mathbf{k}-\mathbf{q}} \right) - f_{\mathbf{k}-\mathbf{q}} f_{\mathbf{k}'+\mathbf{q}} \left(1 - f_{\mathbf{k}'} \right) \left(1 - f_{\mathbf{k}} \right) \right] \tag{7}$$

$$-V_{\mathbf{k}'-\mathbf{k}+\mathbf{q}}\Big[f_{\mathbf{k}}f_{\mathbf{k}'}\left(1-f_{\mathbf{k}'+\mathbf{q}}\right)\left(1-f_{\mathbf{k}-\mathbf{q}}\right)-f_{\mathbf{k}-\mathbf{q}}f_{\mathbf{k}'+\mathbf{q}}\left(1-f_{\mathbf{k}'}\right)\left(1-f_{\mathbf{k}}\right)\Big] \quad (8)$$

$$+2V_{\mathbf{q}}\Big[\left(f_{\mathbf{k}}-f_{\mathbf{k}-\mathbf{q}}\right)\sum_{\mathbf{k}''}\bar{K}_{\mathbf{k}'',\mathbf{k}',\mathbf{q}}+\left(f_{\mathbf{k}'}-f_{\mathbf{k}'+\mathbf{q}}\right)\sum_{\mathbf{k}''}\bar{K}_{\mathbf{k},\mathbf{k}'',\mathbf{q}}\Big] \quad (9)$$

$$-V_{\mathbf{k}'-\mathbf{k}+\mathbf{q}}\Big[\left(f_{\mathbf{k}}-f_{\mathbf{k}'+\mathbf{q}}\right)\sum_{\mathbf{k}''}\bar{K}_{\mathbf{k}'',\mathbf{k}',\mathbf{k}-\mathbf{k}'-\mathbf{q}}+\left(f_{\mathbf{k}'}-f_{\mathbf{k}-\mathbf{q}}\right)\sum_{\mathbf{k}''}\bar{K}_{\mathbf{k},\mathbf{k}'',\mathbf{k}-\mathbf{k}'-\mathbf{q}}\Big]\,, \quad (10)$$

where we have introduced the energies renormalized by the exchange energy according to $\mathscr{E}_{\mathbf{k}} := \varepsilon_{\mathbf{k}} - \sum_{\mathbf{q}} V_{\mathbf{q}} f_{\mathbf{k}-\mathbf{q}}$. The dynamical variables are directly related to the system energies. By introducing the mean kinetic, exchange, and correlation energy per particle as

$$E_{\mathrm{kin}} := 2n^{-1}\sum_{\mathbf{k}}\varepsilon_{\mathbf{k}}f_{\mathbf{k}}\,, \quad (11)$$

$$E_{\mathrm{ex}} := -n^{-1}\sum_{\mathbf{k},\mathbf{q}}V_{\mathbf{q}}f_{\mathbf{k}}f_{\mathbf{k}-\mathbf{q}}\,, \quad (12)$$

$$E_{\mathrm{corr}} := n^{-1}\sum_{\mathbf{k},\mathbf{k}',\mathbf{q}}V_{\mathbf{q}}\bar{K}_{\mathbf{k},\mathbf{k}',\mathbf{q}}\,, \quad (13)$$

respectively, the total energy is given by $\langle H\rangle = n(E_{\mathrm{kin}}+E_{\mathrm{ex}}+E_{\mathrm{corr}})$ with the density $n = 2\sum_{\mathbf{k}} f_{\mathbf{k}}$.

From Eq. (6) - (10) two well-known approximations can be derived: The *Born approximation* (BA) which describes scattering by the bare potential is obtained by retaining only parts (6) and (7). The *random phase approximation* (RPA) consists in keeping the terms (6), (7), and (9) [10, 14]. It accounts for scattering by a dynamically screened potential. Obviously the terms (8) and (10) describe exchange corrections to the terms (7) and (9), respectively, which are necessary to satisfy the correct antisymmetry of the two-particle density matrix upon exchange of the final or initial states. This antisymmetry is violated in the cases BA and RPA. We will discuss the role of exchange contributions for the dynamics by comparing BA and RPA results with calculations comprising the respective exchange terms denoted by BAX [parts (6) - (8)] and RPAX [parts (6) - (10)].

According to Eqs. (5) - (10) relaxation can be identified as a two step process: first, the correlation is built up from the distribution function and, second, the distribution function changes due to the feedback from the correlation. The screening terms, on the other hand, already require the existence of a correlation. This is the reason for the retarded build-up of screening. The screening at a given time t does not only depend on the distribution function at the same time but also on its history; screening much like scattering is a non-Markovian process in a quantum kinetic treatment.

To clearly identify this build-up of screening we will compare the results of the four levels of the theory introduced above with the case where the fully dynamical and non-Markovian treatment of the screening is replaced by a statically screened Coulomb potential in the Born approximation. Here, the screening is described by the usual Lindhard dielectric function in the static limit,

$$\varepsilon(\mathbf{q},\omega=0) = 1 - V_{\mathbf{q}}\sum_{\mathbf{k},\sigma}\frac{f^{\sigma}_{\mathbf{k}}-f^{\sigma}_{\mathbf{k}-\mathbf{q}}}{\varepsilon_{\mathbf{k}}-\varepsilon_{\mathbf{k}-\mathbf{q}}}. \quad (14)$$

This level of the theory will be denoted by BAL. A particular feature in the present case of a two-dimensional electron gas is the fact that in the long-wavelength limit the Lindhard screening only depends on the distribution function at the band minimum $f_{\mathbf{k}=0}$. We will come back to this point below when discussing the results.

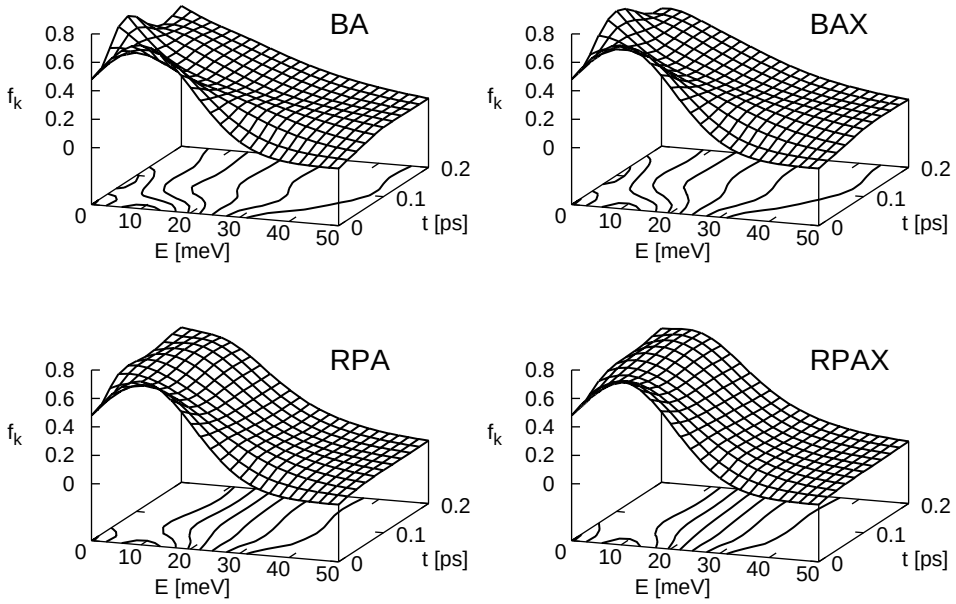

Figure 1. Electron distribution function as a function of the kinetic energy and time for four different levels of the theory as explained in the text. The initial distribution is centered at an excess energy of 10 meV with a width of 15 meV, the density is 7.6×10^{11} cm^{-2}.

3. Results

We apply the theory to the case of the relaxation of a given initial distribution which is assumed to be uncorrelated. The initial distribution is taken to be Gaussian in energy which mimics a carrier distribution generated by an ultrashort laser pulse. Figure 1 displays the relaxation dynamics of an initial distribution centered at an excess energy of 10 meV with a width of 15 meV corresponding to a density of 7.6×10^{11} cm^{-2}. In the case of the Born approximation (BA) we find a very fast dynamics associated even with a strong oscillatory contribution in particular at the bottom of the band. The relaxation is somewhat reduced and the oscillations are less pronounced if, in addition to the direct scattering terms, also the exchange terms are included (BAX). If, on the other hand, screening is taken into account on the level of the random phase approximation (RPA) the slowing down of the relaxation dynamics is much more pronounced and the oscillations are almost gone. Also in this case adding exchange contributions (RPAX) leads again to a further reduction of the relaxation.

In all cases the initial non-equilibrium distribution relaxes towards a Fermi-like distribution. As is clearly seen in the figure the effective temperatures of the distribution functions at later times are very different in the four cases. BA corresponds to the highest temperature, in BAX it is slightly reduced and in the two cases including screening (RPA and RPAX) the temperatures are much smaller than in the previous cases. The physical reason for this different behavior is the strongly different degree of correlations. Being a conservative system, the sum of the three energies introduced in Eqs. (11)-(13) is constant. The exchange energy turns out to be approximately the same in all four cases. The absolute value of the correlation energy, however, is much bigger in the cases without screening (BA and BAX) than in those with screening (RPA and RPAX) because in the latter cases the interaction strength is effectively reduced. Since the correlation energy is negative, this implies also a

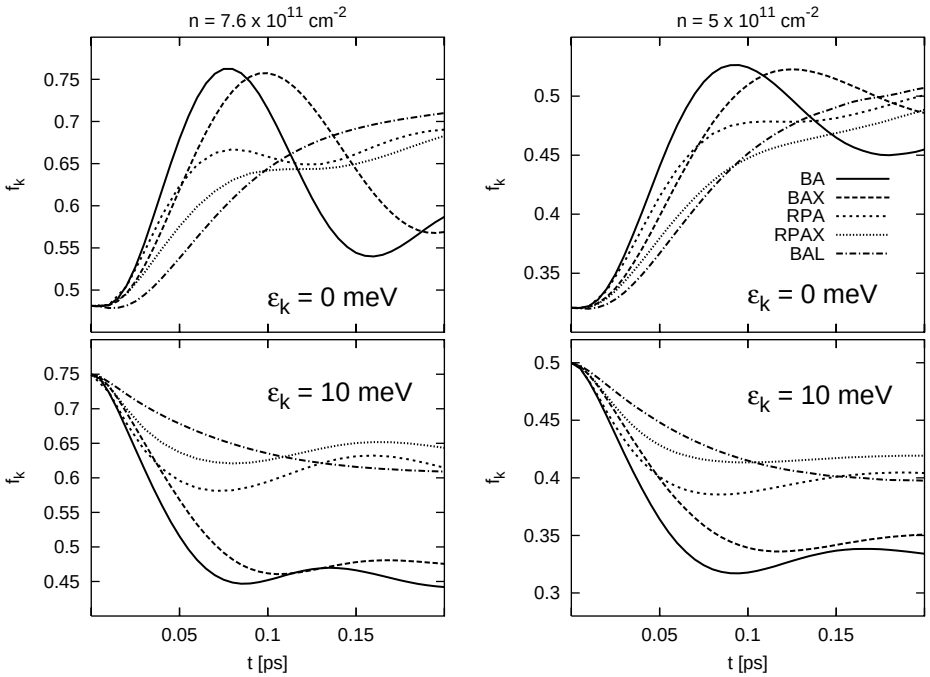

Figure 2. Temporal evolution of the electron distribution function at the minimum of the band (upper part) and at the maximum of the initial distribution (lower part) for the five cases explained in the text. The initial distributions are centered at an excess energy of 10 meV with a width of 15 meV, the density is 7.6×10^{11} cm^{-2} (left column) and 5×10^{11} cm^{-2} (right column).

much bigger kinetic energy in the cases without screening and, thus, an increased effective temperature.

To obtain a more quantitative picture of the relaxation dynamics, in Fig. 2 we have plotted the temporal evolution of the electron distribution at two specific values of **k**, namely at the bottom of the band (upper part) and at the maximum of the initial distribution (lower part). The left column refers to the same initial condition as discussed previously while the right column refers to a reduced density of 5×10^{11} cm^{-2}. Besides the four levels of the theory discussed above, here we have also included the Born approximation with a static Lindhard screening (BAL). The main features are the same at both densities. As can be expected the dynamics is faster in the case of the higher density. In particular the oscillatory behavior at the band minimum in the cases without screening is much more pronounced at the higher density. It turns out that a further lowering of the density results in a complete loss of these oscillations.

Let us now come to a detailed comparison of the dynamics in the five cases studied. A comparison of BA with BAX reveals that the exchange scattering contribution (8) leads to a pronounced reduction of the scattering efficiency especially at early times due to the opposite signs of direct and exchange terms. Since scattering processes are effective just from the beginning the corresponding curves separate from each other immediately and already at 25 fs noticeable differences are seen.

At very early times RPA and RPAX coincide with BA and BAX, respectively. Then, however, the screening builds up and the relaxation of the distribution function dramatically decreases. After about 40-50 fs at the higher density and 50-70 fs at the lower density this reduction due to screening starts to dominate over the reduction by the exchange scattering term (8) as can be seen from the crossing of the curves BAX and RPA in Fig. 2. When

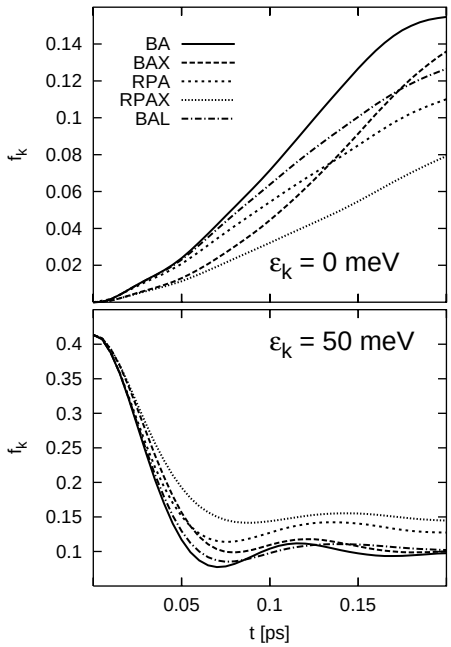

Figure 3. Temporal evolution of the electron distribution function at the minimum of the band (upper part) and at the maximum of the initial distribution (lower part) for the five cases explained in the text. The initial distribution is centered at an excess energy of 50 meV with a width of 15 meV, the density is 5×10^{11} cm^{-2}.

comparing RPAX with RPA we find at early times essentially the same reduction of the relaxation due to the exchange contribution as in the case without screening. At later times (about 120 fs for the higher density and about 150 fs for the lower density) the time evolutions of the distribution functions obtained by RPA and RPAX become more or less parallel. This can be interpreted as a mutual cancellation of the exchange contributions (8) and (10). Thus, at later times the dynamics is well described by the commonly used RPA.

By completely neglecting the retarded build-up of screening, the statically screened model (BAL) strongly overestimates the screening and consequently underestimates the relaxation at short times. For both densities the corresponding curves exhibit a much slower temporal evolution at early times than the other four cases. However, at later times of the order of 200 fs BAL leads to similar results as RPA/RPAX which means that the dynamics can now be described quite well by a non-retarded screening model and obviously also the static limit of the Lindhard dielectric function provides a good approximation.

The situation, however, changes if the initial distribution is characterized by a higher excess energy. This is clearly seen in Fig. 3 where the temporal evolution of the electron distribution function at the bottom of the band and at the maximum of the initial distribution is shown for an excess energy of 50 meV. The general features discussed above for the cases BA/BAX and RPA/RPAX remain the same. Also here, initially RPA coincides with BA and RPAX coincides with BAX demonstrating the retarded build-up of screening and again the relaxation is reduced by the exchange contributions. The static Lindhard screening (BAL), however, now strongly underestimates the screening in contrast to the previous cases where we have found an overestimation of the screening. The reason for this different behavior is the fact that, as mentioned above, the long-wavelength limit of the static screening, which is the most important part, in a two-dimensional electron gas only depends on the distribution function at

the band minimum. In the present case of an excess energy of 50 meV and a width of 15 meV the occupation at $\mathbf{k} = 0$ vanishes initially and, therefore, there is no screening. Indeed, up to about 50 fs the BAL curves coincide with the BA curves and only after the creation of a noticeable occupation at this point screening reduces the relaxation and the curves start to separate. This overestimation of scattering rates by static screening is in qualitative agreement with previous findings where, for a three-dimensional electron gas and in the Boltzmann limit, scattering rates including static and dynamical screening have been calculated for the case of strongly athermal electron distributions [18].

4. Conclusions

In this contribution we have shown that on an ultrashort time scale the inclusion of quantum mechanical exchange terms results in a significant reduction of the scattering efficiency when compared to the case of direct interaction only. With increasing time the screening of the bare Coulomb interaction builds up, but also the screening is reduced by exchange contributions. When the screening has built up, the exchange contributions to scattering and screening tend to compensate each other and the subsequent relaxation dynamics is in good agreement with the standard RPA case. For excess energies which are sufficiently low such that states at the band minimum are occupied the relaxation dynamics at later times can satisfactorily be described in terms of a static screening model while for higher excess energies the static limit of the Lindhard dielectric function strongly underestimates the screening.

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