Enhanced carrier scattering rates in dilute magnetic semiconductors with correlated impurities

F. V. Kryuchenko and C. A. Ullrich

Department of Physics and Astronomy, University of Missouri, Columbia, Missouri 65211

(Dated: May 6, 2018)

In III-V dilute magnetic semiconductors (DMSs) such as Ga$_{1-x}$Mn$_x$As, the impurity positions tend to be correlated, which can drastically affect the electronic transport properties of these materials. Within the memory function formalism we have derived a general expression for the current relaxation kernel in spin and charge disordered media and have calculated spin and charge scattering rates in the weak-disorder limit. Using a simple model for magnetic impurity clustering, we find a significant enhancement of the charge scattering. The enhancement is sensitive to cluster parameters and may be controllable through post-growth annealing.

PACS numbers: 72.80Ey, 78.30Ly

The perspective of utilizing charge and spin of the electrons for new electronic device applications has generated tremendous interest in the field of spintronics [1]. A unique combination of magnetic and semiconducting properties makes dilute magnetic semiconductors (DMSs) very attractive for various spintronics applications [2]. Among the family of DMSs, much attention has been paid to Ga$_{1-x}$Mn$_x$As since the discovery of its relatively high ferromagnetic transition temperature [3], with a current record of $T_c = 159$ K [4].

In Ga$_{1-x}$Mn$_x$As, unlike in II-VI DMSs, the magnetic ions in substitutional positions act as acceptors delivering one hole per ion. All Ga$_{1-x}$Mn$_x$As samples are, however, heavily compensated, with hole concentrations much less than $x$. This signals the presence of substantial amounts of donor defects like arsenic antisites As$_{Ga}$ or interstitial manganese ions Mn$_{II}$ generated during low temperature molecular beam epitaxial growth [4]. The magnetic and transport properties of Ga$_{1-x}$Mn$_x$As depend not only on the manganese fraction $x$ but are extremely sensitive to detailed growth conditions [5], as well as to temperature and speed of post-growth annealing [6, 7, 8]. This sensitivity points to the crucial role played by the defects and their configuration, and has stimulated intense research on the structure of defects and their influence on the magnetic and transport properties of DMSs [2, 9, 10].

Most theoretical models for transport in DMSs assume random defect distributions. However, the presence of both positively and negatively charged defects results in a correlation of their positions. Indeed, Timm et al. [2] found in the limit of thermal equilibrium that, driven by Coulomb attraction, the defects tend to form clusters. The main effect of clustering is ionic screening of the disorder Coulomb potential, which has been shown to be necessary to correctly reproduce the band gap, metal-insulator transition and shape of the magnetization curve [3].

In this paper, we demonstrate that the correlation of defect positions can have a dramatic effect on electronic transport in DMSs: the conductivity of Ga$_{1-x}$Mn$_x$As is strongly modified through a momentum dependent impurity structure factor. We will show that the clustering significantly increases the charge scattering relaxation rate. At the same time, positional correlation taken alone is not sufficient to affect spin scattering: orientational correlation of spin scatterers is also necessary.

To describe the transport properties of DMSs we have employed the memory function formalism [11, 12, 13]. The central point of this approach is the calculation of the current relaxation kernel (or memory function), whose imaginary part can be associated with the Drude relaxation rate. To get an expression for the memory function in spin- and charge-disordered media we have used the equation of motion approach [14, 15] (details of the derivation will be presented elsewhere). Here, we are particularly interested in a paramagnetic system in the long-wavelength limit, the case relevant for studying the conductivity in DMSs above $T_c$. In this case the memory function is obtained as

$$M(\omega) = \frac{V^2}{nm\omega} \sum_{i,\mu} \langle \hat{U}_{\mu}(-\mathbf{k}) \hat{U}_{\nu}(\mathbf{k}) \rangle_{H_m} \times \left( \chi^{\mu\nu}_{\rho\sigma}(\mathbf{k}, \omega) - \chi^{\mu\nu}_{\rho\sigma}(\mathbf{k}, 0) \right),$$

(1)

where $n$ is the carrier concentration and the operators $\hat{U}^\mu$ are defined through a four-component $(\mu = 1, +, -, \tau)$ charge and spin density vector,

$$\hat{U}^\mu(\mathbf{k}) = \frac{1}{V} \sum_{\mathbf{q}} \sum_{\tau, \tau'} (\sigma^\mu)_{\tau \tau'} \hat{\rho}^c_{q, \tau} \hat{\sigma}_{\mathbf{k} - \mathbf{q}, \tau}.$$  

(2)

Here, $\sigma^\mu$ is defined via the Pauli matrices, where $\sigma^1$ is the $2 \times 2$ unit matrix, $\sigma^\pm = (\sigma^x \pm i\sigma^y)/2$, and $\chi^{\mu\nu}_{\rho\sigma}(\mathbf{k}, \omega)$ are the associated charge- and spin-density response functions. The superscript $c$ in Eq. (1) refers to a clean (defect-free) system.

The presence of impurities, including their correlations, enters in Eq. (1) through the expression $\langle \hat{U}_{\mu}(-\mathbf{k}) \hat{U}_{\nu}(\mathbf{k}) \rangle_{H_m}$. The angular brackets indicate a thermodynamical average with respect to a magnetic subsystem Hamiltonian $H_m$. We assume $H_m$ to be a sum of individual spin contributions corresponding to uncorrelated and noninteracting localized spins. The disorder scattering potential is described by the four-component impurity charge- and spin-density operator

$$\hat{U}(\mathbf{k}) = \frac{1}{V} \sum_j \left( \frac{U_j(\mathbf{k})}{2} \hat{S}_j^z \hat{S}_j^z \right) e^{i\mathbf{k} \cdot \mathbf{R}_j},$$

(3)

where the summation is performed over all defect positions. In order to separate the effect of the impurity structure factor from other effects of clustering like ionic screening we
of randomness is a characteristic feature of spin scattering. In
girdless of spatial correlations. The presence of a macroscopic magnetization is not necessary
hibit terms of scattering, localized spins are correlated if they ex-
the presence of a macroscopic magnetization is not necessary
stitutional positions. All defects in Eq. (3) produce thus the
able of frozen spins. For the charge component \( U_1(k) \) we take a
coulomb potential screened with the host material dielectric constant. Effects of ionic screening are disregarded, and
from the electron liquid is absorbed in the response functions.
Let us now consider the correlated product of two compo-
nents of the disorder potential in Eq. (1). First, we separate
first term on the right-hand side of Eq. (4), while the second term contributes only if there are correlations in the
impurity positions. For spin scattering, however, spatial correlations are not sufficient for the pair term to sur-
indeed, if any of the indices \( \mu, \nu \) corresponds to a spin component (e.g., \( \mu = z \)), then the second term in Eq. (4) is
proportional to the average spin and vanishes if \( \langle \hat{S}_z \rangle = 0 \), regard-
less of spatial correlations. The presence of two sources of
randomness is a characteristic feature of spin scattering. In
terms of scattering, localized spins are correlated if they exhibit both
positional and orientational correlations. Note that the presence of a macroscopic magnetization is not necessary for
spins to be correlated. What counts in the scattering is the
short-range orientational correlation that might be present
even in a macroscopically paramagnetic system.
For the charge-scattering term (\( \mu, \nu = 1 \)), we have
\[
\left\langle \hat{U}_1(-k) \hat{U}_1(k) \right\rangle_{\text{H}_m} = |U_1(k)|^2 \frac{n_i}{V} S(k),
\]
with the structure factor
\[
S(k) = 1 + \frac{\Omega_0}{V_x} \sum_{j \neq j'} e^{i k \cdot (R_{j'} - R_j)},
\]
where \( x \) is the molar fraction of magnetic ions in the sample
and \( \Omega_0 \) is the elementary cell volume. Let us introduce a pair
distribution function \( P(R) \), normalized as
\[
\frac{1}{V} \int_V P(R) dR = x,
\]
which describes the probability to find another magnetic ion
at a distance \( R \) from a given ion. We approximate \( S(k) \) as
\[
S(k) \approx 1 + \frac{1}{\Omega_0} \int_V P(R) \cos(k \cdot R) dR.
\]
For a random impurity distribution one has \( P(R) = x \), and
the second term in Eq. (8) vanishes. The structure factor \( S(k) \)
is then equal to 1, which implies a contribution only of the
same-ion term in Eq. (4).
To study the effect of correlations in the defect positions
(higher probability to find magnetic impurities close to each
other), we employ a simple model expression for a pair dis-
tribution function \( P(R) \), assuming it to be a piecewise constant,
spatially symmetrical function of the form
\[
P(R) = \begin{cases} 
  x_c, & R < R_c, \\
  x_d, & R_c < R < R_d \\
  x, & R > R_d .
\end{cases}
\]
(9)
The first region corresponds to a cluster of radius \( R_c \) with
effective impurity concentration \( x_c > x \), the second region is a
depletion layer with \( x_c < x \), necessary to preserve the
average impurity concentration in the sample. The width of
the depletion layer is determined by the normalization condition
\( \int_0^\infty P(R) dR = x_c + R_c x = N \).
The remaining two parameters, \( R_c \) and \( x_c \), describe the
cluster structure and are in general independent. We can re-
late them, however, if we fix the average number \( N \) of the
impurity ions within the cluster:
\[
\frac{4 \pi R_c^3}{3 \Omega_0} x_c = N.
\]
(10)
This seems reasonable for modeling the effect of annealing
on low-temperature grown DMS samples, where one may as-
sume that the total number of Mn\(^{2+}\) substitutional ions within
the cluster is conserved while the cluster size (and density) may vary. In the following we will use \( N = 10 \), consistent
with the results of Monte-Carlo calculations [9].
Our model pair correlation function \( S(k) \) yields the following
momentum-dependent impurity structure factor:
\[
S(k) = 1 + \frac{2 \pi x}{k^3 \Omega_0} \left\{ \frac{2 x_c - x}{x} \left[ \sin(kR_c) - kR_c \cos(kR_c) \right] \right. \\
- \left. \left[ \sin(kR_d) - kR_d \cos(kR_d) \right] \right\}.
\]
(11)
Fig. 1 shows \( S(k) \) for two different values of the cluster radius
\( R_c \), which in principle can be controlled by annealing. As ex-
pected, the structure factor oscillates with decreasing amplit-
ude, with a larger first maximum for smaller cluster size.
Eq. (11) contains the set of full system charge- and spin-
density response functions and, strictly speaking, should
be calculated by iterations. This approach was realized in
(12) to study a spin-independent system close to the metal
insulator threshold. In our case, however, we assume that the
disorder is weak enough so we can approximate Eq. (11)
by expanding to second order in the disorder potential \( \hat{U}(k) \), and thus replace \( \chi_{\mu \nu}^{\rho \rho'}(\mathbf{k}, \omega) \) by its clean system counterpart \( \chi_{\mu \nu}^{\rho \rho'}(\mathbf{k}, \omega) \).
An accurate description of carrier-mediated ferromagnetism
[17] and optical response [18] of Ga\(_{1-x}\)Mn\(_x\)As would
require taking the true multiband structure of the material into
density-density response function of the interacting system as

\[ \chi_{nn}(q, \omega) = \frac{\chi_0(q, \omega)}{\epsilon_{\text{RPA}}(q, 0)}, \] (12)

where \( \chi_0(q, \omega) \) is the non-interacting density-density response function of the clean system, i.e., the Lindhard function, and \( \epsilon_{\text{RPA}}(q, 0) \) is the static RPA dielectric function [19]. The spin response functions in the paramagnetic state are not affected by electron-electron interactions on the RPA level, and they can also be expressed in terms of the Lindhard function \( \chi_0(q, \omega) \).

Under the above approximations Eq. (11) can be directly evaluated and the memory function becomes the sum of spin and charge contributions

\[ M(\omega) = \frac{1}{\tau_n} + \frac{1}{\tau_s}, \] (13)

with

\[ \frac{1}{\tau_n(\omega)} = A \int_0^\infty k^4 S(k) \left| U_1(k) \right|^2 \frac{\chi_0(k, \omega) - \chi_0(k)}{\omega} dk, \] (14)

and

\[ \frac{1}{\tau_s(\omega)} = A \frac{J^2}{4} S_{\text{Mn}}(S_{\text{Mn}} + 1) \int_0^\infty k^4 \frac{\chi_0(k, \omega) - \chi_0(k)}{\omega} dk, \] (15)

where \( S_{\text{Mn}} = 5/2 \) is the localized spin of magnetic impurities, and the common prefactor is given by \( A = (n_i/n)V^2/6\pi^2m. \)

The imaginary parts of Eqs. (14) and (15) represent the energy dependent charge- and spin-scattering contributions to the Drude relaxation rate. Generally speaking, within our model the relaxation times \( \tau_n(\omega), \tau_s(\omega) \) are also momentum dependent. In the present paper, however, we consider only a long-wavelength limit, setting \( q \to 0. \) Note that the momentum-dependent impurity structure factor \( S(k) \) does not appear in the spin term (15).

We have evaluated the scattering rates, Eqs. (14) and (15), for the case of \( \text{Ga}_{0.95}\text{Mn}_{0.05}\text{As} \) with a hole concentration of \( p = 0.5 \) hole per magnetic ion. Other parameters used are: heavy hole effective mass \( m = 0.5\, m_0, \) dielectric constant \( \varepsilon = 13, \) and exchange constant \( VJ = 55\, \text{meV}\, \text{nm}^3, \) which corresponds to the widely used DMS p-d exchange constant \( N_0/\beta = 1.2\, \text{eV}^{1/2}. \)

In Fig. 1 we plot the frequency dependence of the imaginary part of Eqs. (14) and (15). Both spin and charge relaxation rates demonstrate the frequency dependencies that one might expect for momentum-independent scattering and for Coulomb scattering. In agreement with earlier estimations by golden rule [20], the charge scattering in dc limit dominates the spin scattering. We would like to point out, however, that in our calculations (as well as in Ref. [20]) we did not take into account the effect of ionic screening shown [20] to reduce significantly the charge disorder potential. In any case, even in the present model the spin scattering contribution is clearly not negligible, and reaches the same order of magnitude as charge scattering for higher frequencies. Both contributions should therefore be taken into account simultaneously.

Correlation in impurity positions results in a significant increase of the charge scattering contribution at low frequency, while for higher \( \omega \) this enhancement decreases. The origin of this effect is illustrated in the inset in Fig. 2. The presence of impurity clusters selects excitations within a finite momentum window defined by the impurity structure factor. On the other
hand, the region of one-particle excitations is given by the imaginary part of the Lindhard function $\chi_{0}$ (shaded region in Fig. 2). The maximum enhancement of the relaxation rate corresponds to maximum overlap of the selection window with the one-particle spectrum, which happens at low frequency. For higher $\omega$ the window falls outside of the available excitation spectrum, reducing the relaxation rate enhancement.

In Fig. 3 we plot the cluster enhancement factor

$$\xi = \frac{\tau_{n}^{-1} + \tau_{s}^{-1}}{(\tau_{n}^{R})^{-1} + (\tau_{s}^{R})^{-1}}$$

as a function of cluster configuration. The enhancement factor is defined as the ratio of the total (charge plus spin) relaxation rates for correlated and random impurity distributions. Recall that the average number of magnetic ions within the cluster is fixed to $N = 10$, and the cluster radius $R_{c}$ is related to concentration of magnetic ions within the cluster $x_{c}$ through Eq. (10). The enhancement is strongest (up to a factor of 3) for low frequency, and is quite sensitive to cluster configuration. The latter will be sensitive to post-growth annealing, which is widely used to increase $T_{c}$ in Ga$_{1-x}$Mn$_{x}$As samples. The possible modification of transport properties described here should therefore be taken into account along with other effects of annealing like decrease of the number of interstitial Mn ions and increase of carrier concentration.

To summarize, using the memory function formalism we have considered transport in charge and spin disordered media with potential application for DMSs, with particular emphasis on non-randomness of impurity positions in Ga$_{1-x}$Mn$_{x}$As. We have shown that positional correlations alone of the magnetic impurities do not affect spin scattering; orientational correlations are also necessary. For charge scattering, impurity clustering gives rise to a momentum-dependent impurity structure factor which substantially modifies the transport properties of the material, typically leading to 100% enhancements of relaxation rates. These results should give valuable insights into the effects of annealing on low temperature grown DMS samples.

Finally, the discussion in this paper was limited to DMS in the paramagnetic state. However, it is well known that magnetic ordering in DMS can have a dramatic influence on transport properties [21]. Our approach should be well suited to study these effects.

Acknowledgments

This work was supported by DOE Grant No. DE-FG02-05ER46213.