Temperature-dependent resistivity of ferromagnetic $Ga_{1-x}Mn_xAs$: Interplay between impurity scattering and many-body effects

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The static conductivity of the diluted magnetic semiconductor $Ga_{1-x}Mn_xAs$ is calculated using an equation of motion approach for the current response combined with time-dependent density-functional theory to account for Hartree and exchange interactions within the hole gas. We find that the Coulomb scattering off the charged impurities alone is not sufficient to explain the experimentally observed drop in resistivity below the ferromagnetic transition temperature: the often overlooked scattering off the fluctuations of localized spins is shown to play a significant role.

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I. INTRODUCTION

The perspective of utilizing the charge *and* the spin of the electrons for the development of 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 $Ga_{1-x}Mn_xAs$ since the discovery of its relatively high ferromagnetic transition temperature[2] with a current record of $T_c=185$ K.[3]

Unlike most other III-V DMSs, the nature of the itinerant carriers in $Ga_{1-x}Mn_xAs$ is still under debate. It is widely accepted that for low-doped insulating samples the Fermi energy lies in a narrow impurity band. For more heavily doped, high-T_c metallic samples there are strong indications that the impurity band merges with the host semiconductor valence band forming mostly hostlike states at the Fermi energy with some low-energy tail of disorder-related localized states.[4] First-principles calculations[5–7] have so far not been fully conclusive regarding the nature of the itinerant carriers in this regime and further theoretical studies continue to be necessary. The question is thus, in essence, whether the valence-band[8] or impurity-band[9] picture is more adequate to describe the spectrum of experimental observations in $Ga_{1-x}Mn_xAs$.

The purpose of this paper is to present a study which supports the valence-band picture for electronic transport properties and for the optical conductivity in $Ga_{1-x}Mn_xAs$. In this material, unlike in II-VI DMSs, the magnetic ions in substitutional positions act as acceptors delivering holes and producing not only localized spins but also charged defects. We will argue that it is important to treat disorder and many-body effects beyond the simple relaxation time and static screening models which were used in previous theoretical studies.[10–12]

Earlier we developed a theory of transport in charge- and spin-disordered media which combines a multiband $\mathbf{k}\cdot\mathbf{p}$ approach for an accurate description of the valence-band states with a more comprehensive treatment of disorder and electron-electron interaction.[13,14] Our theory is based on an equation of motion approach for the current-current response function[15,16] and has some similarity with the memory function approach.[17–19]

In this paper we apply our formalism to describe the transport properties of $Ga_{1-x}Mn_xAs$ in the static regime. Specifically, we focus on the pronounced drop of the resistivity below T_c which has been observed for optimally annealed metallic samples.[20–23] There exist several earlier studies addressing this and related problems with different theoretical approaches.[24–26] Lopez-Sancho and Brey[24] considered the temperature dependence of the Coulomb scattering off the acceptor centers and found that the carrier relaxation rate is reduced by around 20% in the ferromagnetic phase, consistent with the experimentally observed drop in resistivity.[20] This drop was attributed entirely to the effects associated with the scattering off Coulomb disorder. The main mechanism found to be responsible for the observed drop in resistivity was the change in the semiconductor band structure: during the transition from the paramagnetic to the ferromagnetic state, the giant spin splitting of the energy bands substantially modifies the shape of the Fermi surface, thus altering the possible scattering wave vectors and, consequently, the magnitude of scattering matrix elements.

These findings speak in favor of the valence-band picture of $Ga_{1-x}Mn_xAs$. However, the model of Ref. 24 employed a simplified treatment for the screening of the charge disorder by itinerant carriers, neglecting the exchange part of the electron-electron interaction within the hole liquid. Furthermore, the scattering off the fluctuations of localized spins was ignored. We will show that both effects play an important role in spin-polarized systems and should be included in the valence-band picture model of the itinerant holes in $Ga_{1-x}Mn_xAs$. In fact, our calculations suggest that the previously proposed origin of the resistivity drop in the ferromagnetic phase should be revised: the main reduction in the scattering rate comes from the suppression of the fluctuations of localized spins in the magnetically ordered state.

II. THEORY

To investigate the drop in resistivity below T_c let us first look at the standard expression for the static conductivity obtained from the semiclassical Boltzmann equation,[11]

$$\sigma_{\alpha\beta}=\frac{e^2}{\hbar V}\sum_{n,\mathbf{k}}\frac{\tau_{n,\mathbf{k}}}{\hbar}\frac{\partial E_{n,\mathbf{k}}}{\partial k_\alpha}\frac{\partial E_{n,\mathbf{k}}}{\partial k_\beta}\delta(E_{n,\mathbf{k}}-E_f), \tag{1}$$

where the summation is over the wave vector $\mathbf{k}$ and the energy-band index n. The part of Eq. (1) which is most sen-

sitive to temperature is the carrier scattering rate $\tau_{n,\mathbf{k}}^{-1}$. The task is therefore to derive a microscopic expression for $\tau_{n,\mathbf{k}}^{-1}$ which accounts for all relevant scattering mechanisms, as well as for electronic many-body effects.

We consider a system described by the Hamiltonian

$$\hat{H} = \hat{H}_e + \hat{H}_m + \hat{H}_d, \tag{2}$$

where $\hat{H}_e$ is the contribution of the itinerant carriers and $\hat{H}_m$ represents the subsystem of localized magnetic spins. These two terms constitute the "clean" part of the total Hamiltonian. The last term in Eq. (2) describes disorder in the system

$$\hat{H}_d = V^2 \sum_{\mathbf{k}} \hat{\tilde{\mathcal{U}}}(\mathbf{k}) \cdot \hat{\tilde{\rho}}(-\mathbf{k}), \tag{3}$$

where the four-component charge- and spin-disorder scattering potential

$$\hat{\tilde{\mathcal{U}}}(\mathbf{k}) = \frac{1}{V}\sum_j \begin{pmatrix} U_j(\mathbf{k}) \\ -\frac{J}{2}\hat{S}_j^- \\ -\frac{J}{2}\hat{S}_j^+ \\ -\frac{J}{2}(\hat{S}_j^z - \langle S\rangle) \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{R}_j} \tag{4}$$

is coupled to the four-component vector of charge- and spin-density operators of itinerant carriers

$$\hat{\tilde{\rho}} = \begin{pmatrix} \hat{\rho}^1 \\ \hat{\rho}^+ \\ \hat{\rho}^- \\ \hat{\rho}^z \end{pmatrix} = \begin{pmatrix} \hat{n} \\ \hat{s}^+ \\ \hat{s}^- \\ \hat{s}^z \end{pmatrix} \tag{5}$$

with the components

$$\hat{\rho}^\mu(\mathbf{k}) = \frac{1}{V}\sum_{\mathbf{q}}\sum_{nn'} \langle u_{n',\mathbf{q}-\mathbf{k}}|\sigma^\mu|u_{n,\mathbf{q}}\rangle \hat{a}^+_{n',\mathbf{q}-\mathbf{k}}\hat{a}_{n,\mathbf{q}}. \tag{6}$$

Here, σ^μ $(\mu=1,+,-,z)$ is defined via the Pauli matrices, where σ^1 is the 2×2 unit matrix, $\sigma^\pm=(\sigma^x \pm i\sigma^y)/2$, and $|u_{n,\mathbf{q}}\rangle$ are the two-component Bloch function spinors with wave vector $\mathbf{q}$ and band index n. The summation in Eq. (4) is performed over all defects. Note that the mean-field part of p-d exchange interaction between itinerant holes and localized spins is absorbed into the clean system band-structure Hamiltonian $\hat{H}_e$; disorder in our model consists of the Coulomb potential of charge defects and fluctuations of localized spins around the mean-field value.

The general case of multiple types of defects, including defect correlations, was considered in Ref. 13. For simplicity we here include only the most important defect type, namely, randomly distributed manganese ions in gallium substitutional positions (Mn_{Ga}). Our model treats localized spins as quantum-mechanical operators coupled to the band carriers via a contact Heisenberg interaction featuring a momentum-independent exchange constant J. We use the value of $VJ=-55$ meV nm^3, which corresponds to the widely used DMS p-d exchange constant $N_0\beta=-1.2$ eV.[8] The z axis is chosen along the direction of the macroscopic magnetization.

Our theory of electronic transport in spin- and charge-disordered media is based on the equation of motion[15,16] for the paramagnetic current response of the full, disordered system

$$\chi_{j_{p\alpha}j_{p\beta}}(\mathbf{r},\mathbf{r}',\tau) = -\frac{i}{\hbar}\Theta(\tau)\langle[\hat{j}_{p\alpha}(\tau,\mathbf{r}),\hat{j}_{p\beta}(\mathbf{r}')]\rangle_H, \tag{7}$$

where

$$\hat{j}_{p\alpha}(\tau,\mathbf{r}) = e^{i/\hbar\hat{H}\tau}\hat{j}_{p\alpha}(\mathbf{r})e^{-i/\hbar\hat{H}\tau} \tag{8}$$

is the paramagnetic current-density operator in Heisenberg representation and $\alpha,\beta=x,y,z$ are Cartesian coordinates. During the derivation we assumed our system to be macroscopically homogeneous, which is justified if the coherence length of the electrons is much shorter than the system size. In this case, summing over all electrons will leave us with an averaged effect of disorder that does not depend on the particular disorder configuration. For such macroscopically homogeneous systems the response at point $\mathbf{r}$ depends only on the distance $\mathbf{r}-\mathbf{r}'$ to the perturbation and not on the particular choice of points $\mathbf{r}$ and $\mathbf{r}'$.

A detailed derivation of the time evolution of the response function, Eq. (7), has been carried out in Ref. 14; a key step along the derivation involves the fluctuating force response function

$$\chi_{F_\alpha^d F_\beta^d}(\mathbf{q},\tau) = -\frac{i}{\hbar}\Theta(\tau)\langle[\hat{F}_\alpha^d(\mathbf{q},\tau),\hat{F}_\beta^d(-\mathbf{q})]\rangle_H, \tag{9}$$

where

$$\hat{F}_\alpha^d(\mathbf{q},\tau) = -\frac{iV}{m}\sum_{\mathbf{q}'} q'_\alpha \hat{\tilde{\mathcal{U}}}(\mathbf{q}',\tau)\cdot\hat{\tilde{\rho}}(\mathbf{q}-\mathbf{q}',\tau) \tag{10}$$

is the fluctuating (disorder-induced) part of the driving force

$$\hat{F}_\alpha(\mathbf{q},\tau) = \frac{\partial}{\partial\tau}\hat{j}_{p\alpha}(\mathbf{q},\tau). \tag{11}$$

To deal with the mixing of band carrier and localized spin operators that appears in expression (9), we make the following decoupling approximation

$$\begin{aligned}\langle[\hat{\tilde{\mathcal{U}}}(\mathbf{k},\tau)\cdot\hat{\tilde{\rho}}(\mathbf{q}-\mathbf{k},\tau),\hat{\tilde{\mathcal{U}}}(\mathbf{k}')\cdot\hat{\tilde{\rho}}(-\mathbf{q}-\mathbf{k}')]\rangle_H \\ \approx \sum_{\mu\nu}\langle\hat{\mathcal{U}}_\mu(\mathbf{k},\tau)\hat{\mathcal{U}}_\nu(\mathbf{k}')\rangle_{H_m}\langle[\hat{\rho}_\mu(\mathbf{q}-\mathbf{k},\tau),\hat{\rho}_\nu(-\mathbf{q}-\mathbf{k}')]\rangle_{H_e+H_d} \\ + \langle[\hat{\mathcal{U}}_\mu(\mathbf{k},\tau),\hat{\mathcal{U}}_\nu(\mathbf{k}')]\rangle_{H_m}\langle\hat{\rho}_\nu(-\mathbf{q}-\mathbf{k}')\hat{\rho}_\mu(\mathbf{q}-\mathbf{k},\tau)\rangle_{H_e+H_d}.\end{aligned} \tag{12}$$

This decoupling procedure means that we neglect the influence of the itinerant carriers on the localized spins. Therefore, our approach does not include magnetic polaron effects and lacks the microscopic features of carrier-mediated ferromagnetism. The latter, however, can be reinstated to some

extent by introducing a phenomenological Heisenberg-type term in the magnetic subsystem Hamiltonian $\hat{H}_m$. By comparing the final expression for the current response with the Drude formula in the weak disorder limit we identify the tensor of Drude-type frequency- and momentum-dependent relaxation rates of the form

$$\tau_{\alpha\beta}^{-1}(\mathbf{q},\omega) = i\frac{V^2}{nm\omega}\sum_{\mu\nu}\sum_{\mathbf{k}} k_\alpha k_\beta \langle \hat{\mathcal{U}}_\mu(-\mathbf{k})\hat{\mathcal{U}}_\nu(\mathbf{k})\rangle_{H_m} \times [\chi_{\rho^\mu\rho^\nu}(\mathbf{q}-\mathbf{k},\omega) - \chi^c_{\rho^\mu\rho^\nu}(\mathbf{k},0)] + \tau_A^{-1}, \quad (13)$$

where n is the carrier concentration, $\chi_{\rho^\mu\rho^\nu}(\mathbf{k},\omega)$ are charge- and spin-density response functions associated with the operators, Eq. (6), and the superscript c refers to the clean system. The form of Eq. (13) is similar to the frequency- and momentum-dependent memory function used in the standard memory function formalism.[17–19]

The last term in Eq. (13) stands for additional contributions which arise in magnetically ordered systems only. They result from the second term of Eq. (12) and cannot be expressed in the convenient form of density and spin-density response functions. Here, we just give the clean system and $\mathbf{q}=0$ limit, which suffices to illustrate the general structure

$$\tau_A^{-1} = i\frac{V^2}{nm\omega}\sum_{\mu\nu}\sum_{\mathbf{k}} k_\alpha k_\beta \langle [\hat{\mathcal{U}}_\mu(\mathbf{k}),\hat{\mathcal{U}}_\nu(-\mathbf{k})]\rangle_{H_m}\int_0^\infty d\tau \times \frac{iV}{\hbar}\langle \hat{\rho}^\nu(-\mathbf{k})\hat{\rho}^\mu(\mathbf{k},\tau)\rangle_{H_c}(e^{i\omega\tau}-1)e^{-\eta\tau}. \quad (14)$$

The general expression for τ_A^{-1} as well as more details of the derivation of Eq. (13) can be found in Ref. 14.

All information about the itinerant carriers, including band structure and electron-electron interaction, is contained within the set of charge- and spin-density response functions $\chi_{\rho^\mu\rho^\nu}(\mathbf{k},\omega)$. Strictly speaking, these response functions correspond to the disordered system, and Eq. (13) should be calculated self-consistently.[27] We here assume that the disorder is sufficiently weak so that we can approximate Eq. (13) by expanding to second order in the disorder potential $\hat{\mathcal{U}}(\mathbf{k})$, and thus replace $\chi_{\rho^\mu\rho^\nu}(\mathbf{k},\omega)$ by its clean system counterpart $\chi^c_{\rho^\mu\rho^\nu}(\mathbf{k},\omega)$.

To account for the complexity of the band structure, we use a standard eight-band $\mathbf{k}\cdot\mathbf{p}$ approach with contributions from the remote bands taken up to the second order in the wave vector.[28] The mean-field part of the p-d exchange interaction between itinerant holes and localized spins causes a spin splitting of the bands of the semiconductor host material. Technical details of this multiband linear-response approach will be published elsewhere.

The major advantage of Eq. (13) is that it can be combined in a straightforward manner with time-dependent density-functional theory (TDDFT),[29] which allows us to describe electron-electron interaction effects, including correlations and collective modes, in principle, exactly. In TDDFT the charge- and spin-density response functions of the interacting system are written as follows:[30]

$$\underline{\underline{\chi}}^{-1}(\mathbf{q},\omega) = \underline{\underline{\chi_0}}^{-1}(\mathbf{q},\omega) - \underline{\underline{v}}(q) - \underline{\underline{f_{xc}}}(\mathbf{q},\omega), \quad (15)$$

where $\underline{\underline{\chi_0}}$ denotes the matrix of response functions of the noninteracting system, $\underline{\underline{v}}(q)$ is the Hartree part of the electron-electron interactions, and $\underline{\underline{f_{xc}}}$ is the matrix of exchange and correlation kernels. All quantities in Eq. (15) are 4×4 matrices; according to Eq. (5), the first component is charge, and the other components are spin +, −, and z. As a simplification we use only the exchange part of $\underline{\underline{f_{xc}}}$ and apply the adiabatic local spin-density approximation. The local-field factors for partially spin-polarized system were calculated according to Ref. 31.

Since the mean-field part of the p-d exchange interaction is extracted from the disorder Hamiltonian, the total relaxation rate, Eq. (13), can be separated into contributions associated with Coulomb disorder and with fluctuations of the localized spins. The transverse component (perpendicular to the magnetization) of the relaxation rate tensor in the long wavelength ($\mathbf{q}=0$) and static ($\omega\to0$) limit then has the form

$$\tau_{xx}^{-1} = \tau_c^{-1} + \tau_s^{-1}, \quad (16)$$

where the charge-disorder contribution is

$$\tau_c^{-1} = i\frac{n_i}{n}\frac{V}{m}\lim_{\omega\to0}\frac{1}{\omega}\sum_{\mathbf{k}} k_x^2|U(k)|^2[\chi_{nn}(\mathbf{k},\omega) - \chi^c_{nn}(\mathbf{k},0)] \quad (17)$$

and the contribution from the fluctuations of the localized spins is given by

$$\tau_s^{-1} = i\frac{n_i}{n}\frac{V}{m}\frac{J^2}{4}\lim_{\omega\to0}\frac{1}{\omega}\sum_{\mathbf{k}} k_x^2\{(\langle\hat{S}_z^2\rangle - \langle\hat{S}_z\rangle^2)[\chi_{s^zs^z}(\mathbf{k},\omega) - \chi^c_{s^zs^z}(\mathbf{k},0)] + \langle\hat{S}^-\hat{S}^+\rangle[\chi_{s^+s^-}(\mathbf{k},\omega) - \chi^c_{s^+s^-}(\mathbf{k},0)] + \langle\hat{S}^+\hat{S}^-\rangle[\chi_{s^-s^+}(\mathbf{k},\omega) - \chi^c_{s^-s^+}(\mathbf{k},0)]\} + \tau_A^{-1}. \quad (18)$$

Here, n_i denotes the concentration of Mn_{Ga} defects. $U(k)$ represents the Coulomb potential of a single acceptor center screened by the dielectric constant of the host material, where we take $\varepsilon=13$ for GaAs; the screening by the electron liquid is absorbed in the band-carrier response functions. The angular brackets in Eq. (18) denote the thermodynamic average with respect to the magnetic subsystem Hamiltonian $\hat{H}_m$ in Eq. (13). We assume $\hat{H}_m$ to be a pairwise Heisenberg-type Hamiltonian. In our calculations we use the experimental value of T_c as an input parameter and apply a standard mean-field approach to obtain the temperature dependence of thermodynamically averaged quantities in Eq. (18). Our treatment of spin disorder corresponds to incoherent scattering[32] off uncorrelated spin fluctuations, a mechanism shown to be the dominant contribution in GaMnAs in the ferromagnetic state.[23]

III. RESULTS AND DISCUSSION

Figure 1 presents the temperature dependence of carrier relaxation rates in $Ga_{1-x}Mn_xAs$ obtained within our model. The calculations were performed for a system with Mn con-

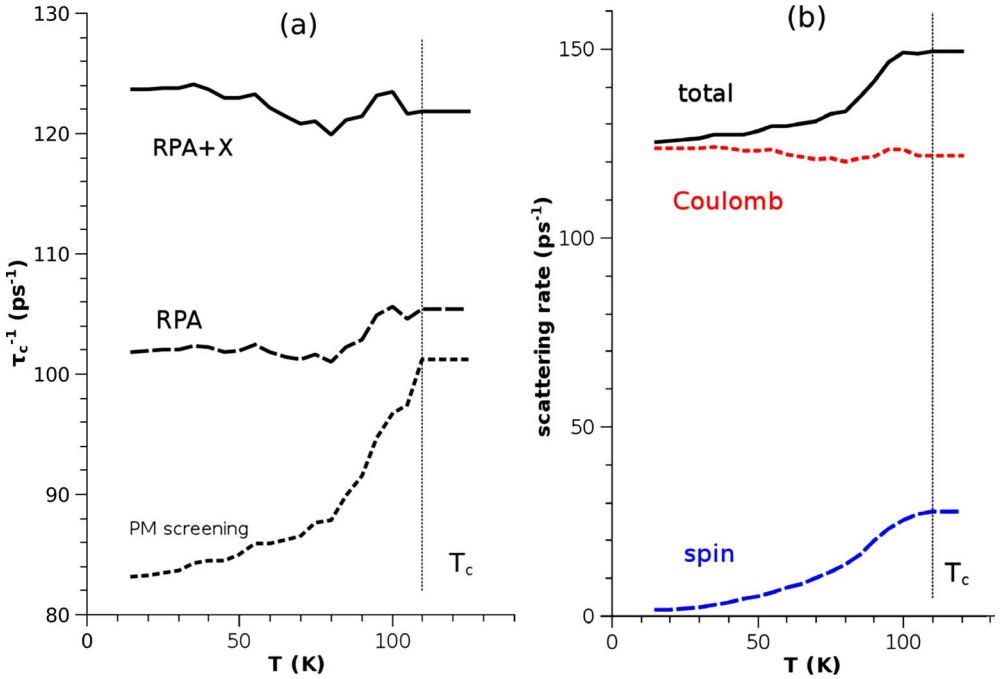

FIG. 1. (Color online) Temperature dependence of the carrier relaxation rates in $Ga_{0.95}Mn_{0.05}As$. The vertical line indicates T_c. (a) Relaxation rates associated with the scattering off Coulomb disorder calculated within different screening models (see text for details). (b) Total relaxation rate (solid line) and contributions from scattering off Coulomb disorder (dotted line) and off localized spin fluctuations (dashed line).

centration $x=0.05$ and carrier concentration of $p=0.3$ holes per Mn in Ga substitutional positions. The left panel shows the contributions from the scattering off Coulomb disorder with screening effects accounted for by three different methods. The dotted line corresponds to screening described within the Thomas-Fermi approximation for paramagnetic systems. The screening here is temperature independent and the 20% drop in the scattering rate in the ferromagnetic phase is entirely due to modification of the possible scattering wave vectors.

If, however, we allow changes in the band structure to affect the screening as well, e.g., on the random-phase approximation (RPA) level [first two terms in Eq. (15) and dashed line in Fig. 1(a)], the drop in the resistivity in the ferromagnetic phase is significantly reduced. This effect was also considered in Ref. 24. But if we now go further and include the exchange part of electron-electron interaction in Eq. (15), then the drop in the resistivity is completely washed out, see the solid line in Fig. 1(a). Moreover, for some parameters, the trend is reversed and the scattering off Coulomb disorder actually *increases* in the ferromagnetic phase.

The explanation for this behavior lies in the exchange part, which counteracts the larger Hartree part of the electron-electron interaction and reduces the screening of the Coulomb disorder potential calculated within RPA. Therefore we have an overall increase in the charge relaxation rate once the exchange part of electron-electron interaction is taken into account, see Fig. 1(a). On the other hand, the exchange part of the electron-electron interaction is more pronounced for spin-polarized systems, resulting in a stronger reduction in the screening of the disorder potential and thus causing an increase in the Coulomb scattering in the ferromagnetic phase. This process competes with and, for some parameters, reverses the reduction in the relaxation rate due to band-structure-related modifications of the scattering wave vectors.

It is thus apparent that the scattering off the Coulomb disorder potential alone cannot be responsible for the experimentally observed drop in resistivity. The other possible contribution is the scattering off the fluctuations of localized spins. In Fig. 1(b) we plot the temperature dependence of the scattering rate for both mechanisms. The scattering off spin fluctuations is often overlooked since its magnitude is substantially smaller than that of the Coulomb scattering. Due to effective suppression of spin fluctuations in the ferromagnetic phase, however, the temperature dependence of this relaxation mechanism is much more pronounced. Indeed, in a fully spin-polarized state, the scattering takes place only off the quantum fluctuations of localized spins. The total relaxation rate, Eq. (16), which is the sum of both contributions, restores its 20% drop during transition from paramagnetic to ferromagnetic phase. The majority of this drop is found to be due to the suppression of the scattering off localized spin fluctuations.

In Fig. 2 we plot the temperature dependence of the static resistivity of $Ga_{0.95}Mn_{0.05}As$, normalized to the paramagnetic state value. The calculation was done according to Eqs. (1) and (16)–(18). Solid and dashed lines correspond to different levels of compensation in the system, 0.3 and 0.5 hole per substitutional Mn, respectively (in practice, this number is difficult to control). The open squares represent the experimental data of Ref. 20. The theory demonstrates good agreement with experiment.

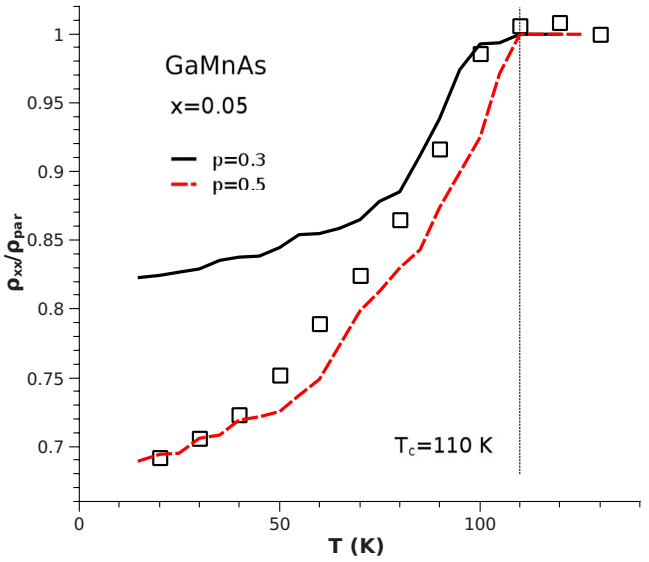

FIG. 2. (Color online) Static resistivity of $Ga_{0.95}Mn_{0.05}As$ normalized to the paramagnetic state value. Solid and dashed lines: compensation levels $p=0.3$ and $p=0.5$ (number of holes per Mn_{Ga}). The vertical line indicates $T_c=110$ K. Experimental points are from Ref. 20.

IV. CONCLUSION

We have developed a theory of transport in spin- and charge-disordered media within the valence-band picture of metallic GaMnAs. The approach combines the multiband $\mathbf{k}\cdot\mathbf{p}$ description of the semiconductor band structure with a microscopic treatment of disorder and dynamical electron-electron interaction by the methods of TDDFT. We illustrate our formalism by describing the experimentally observed drop in static resistivity of GaMnAs in the ferromagnetic phase. This problem had been addressed before in Ref. 24 but with a model lacking some important features such as scattering off the fluctuations of localized spins and electron-electron interactions beyond RPA that can naturally be included in our formalism. Similar to Ref. 24, we obtained agreement with the experimental observations, but the underlying physics is quite different. Much of the drop of resistivity in the ferromagnetic phase is found to be due to the suppression of localized spin fluctuations in the magnetically ordered state.

To conclude, we have developed a theoretical description of itinerant carriers in DMSs within the valence-band picture that accounts for band structure, scattering from Coulomb and magnetic impurities, and screening via dynamical many-body effects. An accurate description of static transport properties in GaMnAs involves a subtle interplay of all these ingredients. Our approach is also suitable for frequency-dependent properties[12,33] such as the optical conductivity, where the presence of collective modes is expected[14] to substantially modify the frequency-dependent carrier relaxation rates. These effects will be discussed elsewhere.

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