Final Report

Charge and Spin Transport in Dilute Magnetic Semiconductors

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Project Summary

This proposal to the DOE outlines a three-year plan of research in theoretical and computational condensed-matter physics, with the aim of developing a microscopic theory for charge and spin dynamics in disordered materials with magnetic impurities. Important representatives of this class of materials are the dilute magnetic semiconductors (DMS), which have attracted great attention as a promising basis for spintronics devices. There is an intense experimental effort underway to study the transport properties of ferromagnetic DMS such as (Ga,Mn)As, and a number of interesting features have emerged: negative magnetoresistance, anomalous Hall effect, non-Drude dynamical conductivity, and resistivity maxima at the Curie temperature. Available theories have been able to account for some of these features, but at present we are still far away from a systematic microscopic understanding of transport in DMS. We propose to address this challenge by developing a theory of charge and spin dynamics based on a combination of the *memory-function formalism* and *time-dependent density functional theory*. This approach will be capable of dealing with two important issues: (a) the strong degree of correlated disorder in DMS, close to the localization transition (which invalidates the usual relaxation-time approximation to the Boltzmann equation), (b) the essentially unknown role of dynamical many-body effects such as spin Coulomb drag. We will calculate static and dynamical conductivities in DMS as functions of magnetic order and carrier density, which will advance our understanding of recent transport and infrared absorption measurements. Furthermore, we will study collective plasmon excitations in DMS (3D, 2D and quantum wells), whose linewidths could constitute a new experimental probe of the correlation of disorder, many-body effects and charge and spin dynamics in these materials.

1 Introductory remarks

The starting date for this grant was June 30, 2004. At this time, the PI was still affiliated with the University of Missouri-Rolla (UMR).[1] On January 1, 2005, the PI transferred from UMR to the University of Missouri-Columbia (UMC). It was agreed upon between the UMR and UMC Physics Departments that the grant would remain at UMR until the end of the Spring 2005 semester. The grant DE-FG02-04ER46151 was then closed on June 30, 2005, and re-opened at UMC on October 1, 2005, with the new grant number DE-FG02-05ER46213. It has been active since then.

This final report should therefore cover, strictly speaking, only the one-year period from June 30, 2004 to June 30, 2005. However, since the original proposal was for a three-year period, and the foundations for most of the work were laid in the first year, we will give in this report a summary of the research accomplishments over the three-year period 2004-2007. This closely follows the section "Results from previous DOE support" in the proposal entitled "Charge and Spin Dynamics in Bulk and Heterostructured Dilute Magnetic Semiconductors" which was submitted to DOE in April 2007 and approved for funding under grant DE-FG02-05ER46213.

2 Research Accomplishments

The research carried out under this grant has resulted in 6 publications, 2 invited and 8 contributed talks, from 2004 to 2007.

2.1 Enhanced carrier scattering in DMS with correlated impurities

In $Ga_{1-x}Mn_xAs$, the magnetic ions in substitutional positions act as acceptors delivering one hole per ion. However, heavy compensation signals the presence of substantial amounts of donor defects like arsenic antisites As_{Ga} or interstitial manganese ions Mn_I. The magnetic and transport properties of $Ga_{1-x}Mn_xAs$ are known to be extremely sensitive to detailed growth conditions, as well as to temperature and speed of post-growth annealing. 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.* found in the limit of thermal equilibrium that, driven by Coulomb attraction, the defects tend to form clusters. This is illustrated in Fig. 1.

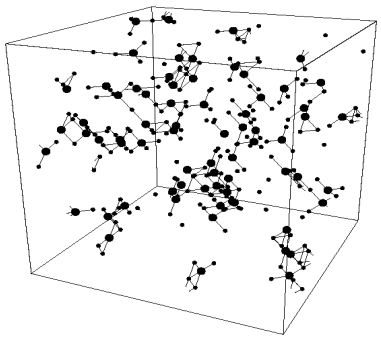

Figure 1: In GaMnAs, Mn antisite and substitutional defects tend to form clusters, simulated with Monte Carlo [From Timm et al.].

We developed a transport theory in charge and spin disordered media with particular emphasis on non-randomness of the impurity positions in $Ga_{1-x}Mn_xAs$. For simplicity, we work in a single-band model with (hole) effective mass m. Starting point is the time evolution of the paramagnetic current-current response function

$$\chi_{j_{p\alpha}j_{p\beta}}(\mathbf{q},t) = -\frac{i}{\hbar}\Theta(t)\langle[\hat{j}_{p\alpha}(\mathbf{q},t),\hat{j}_{p\beta}(-\mathbf{q})]\rangle_H. \quad (1)$$

[1] The University of Missouri-Rolla changed its name in 2008 to Missouri University of Science and Technology.

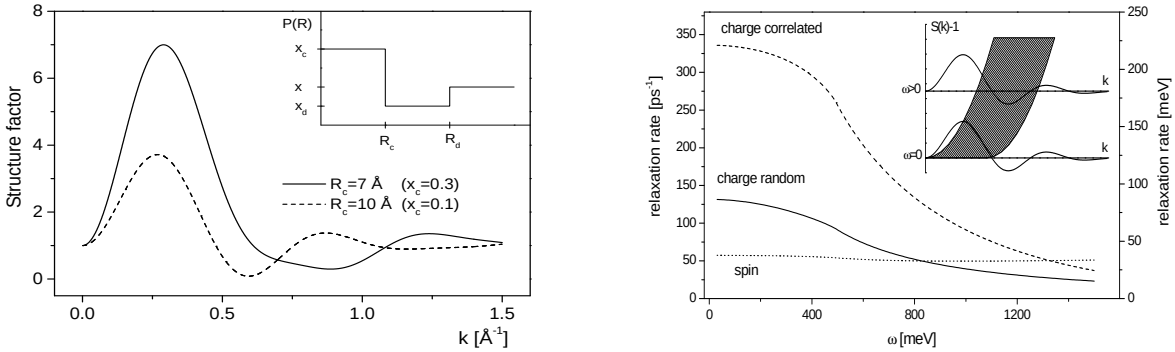

Figure 2: Left: momentum-dependent impurity structure factor for GaMnAs clustering (see inset). Right: charge and spin scattering relaxation rates for random and correlated impurities.

Using an equation of motion approach, it is possible to cast $\chi_{j_{p\alpha}j_{p\beta}}$ into a form where the spin-dependent disorder potential appears explicitly. For a paramagnetic system in the long-wavelength limit, the case relevant for DMSs above T_C, the total current response function is obtained as

$$\chi^J_{\alpha\beta}(\omega) = \chi^c_{j_{p\alpha}j_{p\beta}}(\omega) + \frac{n}{m}\delta_{\alpha\beta} + \frac{V^2}{m^2\omega^2}\sum_{\substack{\mathbf{k}\\ \mu\nu}} k_\alpha k_\beta \left\langle \hat{\mathcal{U}}_\mu(-\mathbf{k})\, \hat{\mathcal{U}}_\nu(\mathbf{k})\right\rangle_{H_m} \Big(\chi_{\rho^\mu\rho^\nu}(\mathbf{k},\omega) - \chi^c_{\rho^\mu\rho^\nu}(\mathbf{k},0)\Big). \quad (2)$$

The operators $\hat{\rho}^\mu$ are based on a four-component ($\mu = 1, +, -, z$) charge and spin density vector,

$$\hat{\rho}^\mu(\mathbf{k}) = \frac{1}{V}\sum_{\mathbf{q}}\sum_{\tau\tau'}(\sigma^\mu)_{\tau\tau'}\, \hat{a}^+_{\mathbf{q}-\mathbf{k},\tau}\, \hat{a}_{\mathbf{q},\tau'}\,. \quad (3)$$

Here, σ^μ 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 $\chi_{\rho^\mu\rho^\nu}(\mathbf{k},\omega)$ are the associated charge- and spin-density response functions. The superscript c in Eq. (2) refers to a clean (impurity-free) system.

Eq. (2) represents a generalization of the *memory function formalism* (MFF). The formal connection between the memory function M and the response function is $\chi(\omega) = \chi_0/[1+\omega/M(\omega)]$. The MFF can thus be viewed as a *generalized relaxation-time approximation*, defining a linear response theory in the presence of extrinsic dissipation.

The presence of impurities, including their correlations, enters in Eq. (2) through the term $\langle\hat{\mathcal{U}}_\mu(-\mathbf{k})\hat{\mathcal{U}}_\nu(\mathbf{k})\rangle_{H_m}$, indicating a thermodynamical average with respect to a magnetic subsystem Hamiltonian $\hat{H}_m$. We assume $\hat{H}_m$ 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, summing over all defect positions:

$$\hat{\mathcal{U}}(\mathbf{k}) = \frac{1}{V}\sum_j \begin{pmatrix} U_1(\mathbf{k}) \\ \frac{J}{2}\hat{S}^j_- \\ \frac{J}{2}\hat{S}^j_+ \\ \frac{J}{2}\hat{S}^j_z \end{pmatrix} e^{i\mathbf{k}\cdot\mathbf{R}_j}\,. \quad (4)$$

We consider only one type of defects, Mn^{2+} ions in cation substitutional positions. All defects in Eq. (4) produce thus the same charge potential and carry localized spins, which are coupled to the band carriers via a Heisenberg exchange interaction. In strongly compensated systems, it is

important to include ionic screening resulting from from Mn_I interstitial defects. To account for this effect, a straightforward generalization of the disorder Hamiltonian has been developed.

To study the effect of correlations in the defect positions, we model a piecewise constant and spherically symmetric pair distribution function $P(\mathbf{R})$ as shown in the inset of Fig. 2. The resulting momentum-dependent impurity structure factor $\mathcal{S}(k)$ is shown in Fig. 2 for two different values of the cluster radius R_c. It is useful to express our results in the familiar terms of transport relaxation times. We cast the conductivity resulting from Eq. (2) into a Drude-like expression in the limit $\omega\tau \gg 1$, which gives the following energy-dependent relaxation times:

$$\frac{1}{\tau_n(\omega)} = A \int_0^\infty k^4 \mathcal{S}(k) \frac{|U_1(k)|^2}{\varepsilon_{\mathrm{RPA}}(k)} \frac{\chi_0(k,\omega) - \chi_0(k)}{-i\omega}\, dk, \tag{5}$$

$$\frac{1}{\tau_s(\omega)} = A\, \frac{J^2}{4}\, S_{\mathrm{Mn}}(S_{\mathrm{Mn}}+1) \int_0^\infty k^4\, \frac{\chi_0(k,\omega) - \chi_0(k)}{-i\omega}\, dk, \tag{6}$$

where $S_{\mathrm{Mn}} = 5/2$ is the localized spin of magnetic impurities, and the common prefactor is given by $A = (n_i/n)V^2/6\pi^2 m$. We evaluate τ_n^{-1} and τ_s^{-1} for $Ga_{0.95}Mn_{0.05}As$ with a hole concentration of $p = 0.5$ per magnetic ion. Other parameters used are: heavy hole effective mass $m = 0.5\, m_0$, dielectric constant $\varepsilon = 13$, and exchange constant $VJ = 55\,\mathrm{meV\,nm^3}$, which corresponds to the widely used DMS p-d exchange constant $N_0\beta = 1.2\,\mathrm{eV}$.

Fig. 2 shows the frequency dependence of the real part of τ_n^{-1} and τ_s^{-1}. In agreement with earlier golden-rule estimates, the charge scattering dominates the spin scattering in the DC limit. The spin scattering contribution is clearly not negligible, and 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 ω this enhancement decreases. 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 $Ga_{1-x}Mn_xAs$ samples.

This work was published in Phys. Rev. B together with Dr. Fedir Kyrychenko.

2.2 Dissipation through spin Coulomb drag in transport and optical excitations

In collaboration with Dr. Irene D'Amico from the University of York (UK), we have studied a critical issue for potential spintronics devices, namely dissipation in spin transport and dynamics due to Spin Coulomb Drag (SCD). The phenomenological expression for the SCD force is given by

$$\mathbf{F}_{\sigma\bar{\sigma}}(\omega) = -\gamma(\omega) m \frac{n_\sigma n_{\bar{\sigma}}}{n} \left(\mathbf{v}_\sigma - \mathbf{v}_{\bar{\sigma}}\right). \tag{7}$$

Here, $\mathbf{F}_{\sigma\bar{\sigma}}$ is the Coulomb force (per unit volume) exerted by spin $\bar{\sigma}(=-\sigma)$ electrons, moving with center-of-mass velocity $\mathbf{v}_{\bar{\sigma}}$, on spin σ electrons, moving with center-of-mass velocity $\mathbf{v}_\sigma$. The number density, n_σ, of σ-spin electrons of effective mass m, and the total density, $n = n_\uparrow + n_\downarrow$, are those of a homogeneous reference system. The drag coefficient $\gamma(\omega, T) = -\frac{ne^2}{m}\, \Re\rho_{\uparrow\downarrow}(\omega, T; n_\uparrow, n_\downarrow)$ appearing in Eq. (7) is directly proportional to the (negative-valued) real part of the spin-transresistivity $\rho_{\uparrow\downarrow}$,

defined by $\mathbf{E}_\uparrow|_{\mathbf{j}_\uparrow=0} = -e\mathbf{j}_\downarrow\rho_{\uparrow\downarrow}$. Here, $\mathbf{j}_\sigma$ is the current density of the σ spin population, and $\mathbf{E}_\sigma$ is the effective electric field coupling to it, including the gradient of the local chemical potential.

The SCD total power loss per unit time for the σ-spin population is given by

$$P_\sigma(\omega) = \int_V d^3r\,[\mathbf{F}_{\sigma\bar{\sigma}}\cdot\mathbf{v}_\sigma] = e^2\int_V d^3r\left[\left(\frac{n_{\bar{\sigma}}(\mathbf{r})}{n_\sigma(\mathbf{r})}\,|\mathbf{j}_\sigma|^2 - \mathbf{j}_{\bar{\sigma}}\cdot\mathbf{j}_\sigma\right)\Re\rho_{\uparrow\downarrow}(\omega,T;n_\uparrow(\mathbf{r}),n_\downarrow(\mathbf{r}))\right]. \qquad (8)$$

P_σ can change sign depending on the relative strength and direction of the spin-resolved current densities, a positive sign implying that the σ spin population is being dragged along by the faster $\bar{\sigma}$ spin population.

In another illustration of the impact of the SCD effect on spin dynamics, we consider intersubband charge- and spin-density excitations (CDE and SDE) in a parabolic quantum well. The inset to Fig. 3 illustrates the two types of plasmons, in which the $n_\uparrow$ and $n_\downarrow$ components move back and forth in phase or with opposite phase. Our derivation of the excitation energies for a spin-dependent system is based on TDDFT. Kohn's theorem predicts that a charge plasmon in a parabolic confining potential is undamped. By contrast, spin plasmons have an intrinsic linewidth due to the SCD.

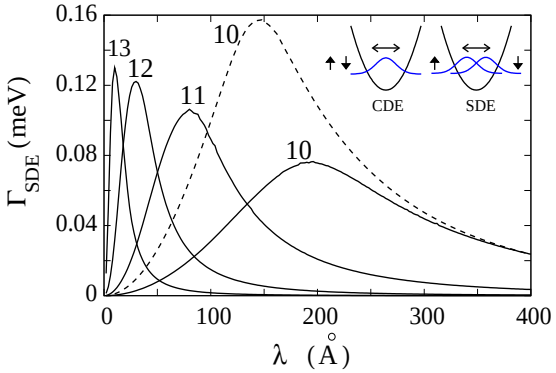

Figure 3: Spin-plasmon linewidth for a parabolic quantum well versus well curvature parameter λ for densities $N_s = 10^x$ cm^{-2}, with x as indicated. Solid lines: InAs, dashed line: GaAs.

The dominant contribution to the intrinsic SDE linewidth Γ for a parabolic quantum well is given by the SCD:

$$\Gamma^{\rm SCD}_{\rm SDE}(\omega) = \frac{e^2N_s\omega}{2\omega^2_{pq\sigma}}\int dz\,\Re\rho_{\uparrow\downarrow}(\omega;n_\uparrow(z),n_\downarrow(z))\left[\frac{n_{\bar{\sigma}}(z)}{n_\sigma(z)}\,|\mathbf{j}_{pq\sigma}(z)|^2 + \mathbf{j}_{pq\bar{\sigma}}(z)\cdot\mathbf{j}_{pq\sigma}(z)\right], \qquad (9)$$

with N_s the two-dimensional electronic sheet density, and $\mathbf{j}_{pq\sigma} = \langle\psi_{p\sigma}|\hat{\mathbf{j}}_\sigma|\psi_{q\sigma}\rangle$, with $\hat{\mathbf{j}}_\sigma$ being the paramagnetic particle current density operator. By comparison with Eq. (8), we immediately recognize the structure of the power loss typical of the Coulomb drag force. Numerical results for $\Gamma^{\rm SCD}_{\rm SDE}$ for GaAs and InAs quantum wells are shown in Fig. 3. Overall, we find that $\Gamma^{\rm SCD}_{\rm SDE}$ can be as high as a large fraction of an meV, thus lying within experimental reach.

Eq. (9) suggests an experiment to extract the impact of SCD on spin dynamics, by an inelastic light scattering measurement of the linewidth of both charge- and spin-plasmons in the *same* parabolic well. Under the reasonable assumption that (i) extrinsic (ext) damping (nonmagnetic impurities, phonons) affect the CDE and SDE in the same way, and (ii) all many-body dissipative effects besides SCD can be disregarded due to the parabolic confinement, we have

$$\Gamma_{\rm SDE} - \Gamma_{\rm CDE} \approx \left(\Gamma^{\rm ext}_{\rm SDE} + \Gamma^{\rm SCD}_{\rm SDE}\right) - \left(\Gamma^{\rm ext}_{\rm CDE}\right) \approx \Gamma^{\rm SCD}_{\rm SDE}\,, \qquad (10)$$

i.e., the SCD contribution to the spin-plasmon linewidth is given to a very good approximation by the difference of the SDE and the CDE linewidths. This provides a valuable opportunity for comparison with microscopic models for the transresistivity via Eq. (9), using the appropriate Kohn-Sham single-particle orbitals of the system.

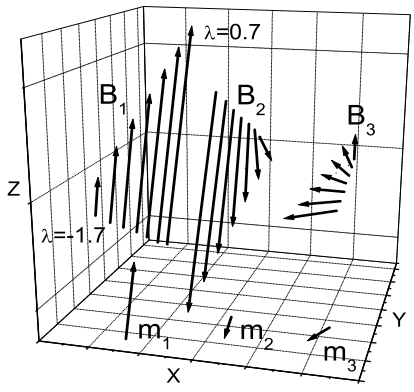

Figure 4: Global noncollinear nonuniqueness for a linear 3-point lattice. All magnetic fields shown here produce the same 4-density.

2.3 Nonuniqueness in spin-DFT on lattices

I have pursued two side projects which address the so-called *nonuniqueness problem* in spin-DFT. At first glance, this problem seems purely formal, but in fact there is a great deal of relevance for the field of spintronics. Among other things, nonuniqueness in DFT has a practical impact on understanding the origins of the spin gap in half metallic ferromagnets, and manipulating qubits by external, non-collinear magnetic fields.

The nonuniqueness refers here to the mapping between 4-densities $\{n(\mathbf{r}), \mathbf{m}(\mathbf{r})\}$ and 4-potentials $\{v(\mathbf{r}), \mathbf{B}(\mathbf{r})\}$. In other words, one can find combinations of scalar potentials and magnetic fields that produce the *same* ground-state density and magnetization. This is in contrast with the non-magnetic case, where the mapping between densities and potentials is unique, thanks to the Hohenberg-Kohn theorem of DFT. Previously, only a few, special cases of nonuniqueness in spin-DFT were known. For example, in a system that is fully spin polarized in $\uparrow$ direction, one can choose the $v_\downarrow$ component of the potential arbitrarily without changing the ferromagnetic ground state (provided there is a gap). However, such statements about nonuniqueness in DFT seem rather trivial and the formal results are too general to be useful in practice.

I have studies the nonuniqueness in spin-DFT on arbitrary *lattices* and discovered two theorems, describing new non-trivial cases of nonuniqueness which I call *local saturation* and *global noncollinear* nonuniqueness. Figure 4 shows an example of global noncollinear nonuniqueness for a linear 3-point lattice. All magnetic fields shown here produce the same ground-state 4-density. This effect has recently been predicted and is here explicitly demonstrated for the first time. The continuum limit of these lattice results is very interesting: it turns out that both new examples become either pathological or exceedingly rare (vanishing mathematical measure), which is somewhat reassuring for the practical applications of spin-DFT. However, if one views discrete lattices such as in Figure 4 as simple models for electrons in coupled quantum dots, it is clear that nonuniqueness is an effect to be reckoned with when attempting qubit operations using magnetic fields.

Another project, in collaboration with Klaus Capelle (Sao Paulo) and Giovanni Vignale (Missouri) studied a subtle loophole in the Hohenberg-Kohn theorem of spin-DFT.

3 Publications and talks resulting from the present grant

3.1 Publications

1. *Degenerate ground states and nonunique potentials: Breakdown and restoration of density functionals.* K. Capelle, C. A. Ullrich, and G. Vignale, Phys. Rev. A **76**, 012508 (2007)

2. *Intrinsic power loss and damping of optical excitations in spintronic devices.* I. D'Amico and C. A. Ullrich, Journal of Magnetism and Magnetic Materials **316**, 484 (2007)

3. *Enhanced carrier scattering rates in dilute magnetic semiconductors with correlated impurities.* F. V. Kyrychenko and C. A. Ullrich, Phys. Rev. B **75**, 045205 (2007)

4. *Dissipation through spin Coulomb drag in electronic spin dynamics and optical excitations.* I. D'Amico and C. A. Ullrich, Phys. Rev. B **74**, 121303(R) (2006); editorially selected for Virt. J. Nanoscale Sci. & Tech. **14**, (13) (2006)

5. *Spin Coulomb drag: an intrinsic dissipation mechanism in spintronics.* I. D'Amico and C. A. Ullrich, Physica Status Solidi (b) **243**, 2285 (2006)

6. *Nonuniqueness in spin-density-functional theory on lattices.*
   C. A. Ullrich, Phys. Rev. B **72**, 073102 (2005)

3.2 Invited talks

1. *Time-dependent density-functional approaches for transport and optical excitations in semiconductors.* 13th Brazilian Workshop on Semiconductor Physics (Sao Paulo, Brazil, April 1-5, 2007)

2. *Microscopic approaches to dissipation in spin-dependent transport and optical excitations in semiconductors.* 53rd Mid-West Solid State Conference (Kansas City, October 8-10, 2006)

3.3 Contributed talks

1. *Electronic transport in diluted magnetic semiconductors: application of the memory function formalism for spin and charge disordered media.* F. V. Kyrychenko and C. A. Ullrich, MRS spring meeting 2007 (San Francisco, April 10-12, 2007)

2. *Electronic transport in diluted magnetic semiconductors: application of the memory function formalism for spin and charge disordered media.* F. V. Kyrychenko and C. A. Ullrich, APS March meeting 2007 (Denver, March 5-9, 2007)

3. *Dissipation through spin Coulomb drag in electronic spin dynamics.* I. D'Amico and C. A. Ullrich, APS March meeting 2006 (Baltimore, March 13-17, 2006)

4. *Electrical conductivity in disordered spin-dependent media: application to diluted magnetic semiconductors* F. V. Kyrychenko and C. A. Ullrich, APS March meeting 2006 (Baltimore, March 13-17, 2006)

5. *Dissipation through spin Coulomb drag in electronic spin dynamics.* I. D'Amico and C. A. Ullrich, 8th International Workshop on Nonlinear Optics and Excitation Kinetics in Semiconductors (Münster, Germany, February 20-24, 2006)

6. *Optical response in disordered spin-dependent media: Application to diluted magnetic semiconductors (Poster).* F. V. Kyrychenko and C. A. Ullrich, 52nd Midwest Solid State Conference (Columbia, Missouri, October 8-9, 2005)

7. *Dissipation through spin Coulomb drag in electronic spin dynamics.* I. D'Amico and C. A. Ullrich, NATO-ASI: "Manipulating Quantum Coherence in Solid State Systems" (Cluj-Napoca, Romania, August 29 -September 8, 2005)

8. *Memory function formalism for the optical response of diluted magnetic semiconductors.* F. V. Kyrychenko and C. A. Ullrich, APS March meeting 2005 (Los Angeles, March 21-25, 2005)

4 Personnel associated with the grant

1. Dr. Carsten A. Ullrich, Associate Professor of Physics

2. Dr. Fedir V. Kyrychenko, Postdoctoral Research Associate