Temperature Dependence of the Dielectric Constant and Resistivity of Diluted Magnetic Semiconductors.

M.P. López-Sanchez and L. Brey.

Instituto de Ciencia de Materiales de Madrid (CSIC), Cantoblanco, 28049, Madrid, Spain.

We study the effect that the ferromagnetic order has on the electrical properties of Diluted Magnetic Semiconductors. We analyze the temperature dependence of the dielectric constant and of the resistivity of Ga$_{1-x}$Mn$_x$As. In our treatment the electronic structure of the semiconductor is described by a six band Kohn-Luttinger Hamiltonian, the thermal fluctuations of the Mn magnetic moments are treated in the mean field approximation, the carrier-carrier interaction within the random phase approximation, and the transport properties using the relaxation time approximation. We find that the Thomas-Fermi length changes near 8% when going from the ferromagnetic to the paramagnetic phase. We also find, in good agreement with the experiments, that the resistivity changes near 20% when going from zero to the Curie temperature. We explain this change in the resistivity in terms of the variation of the Fermi surface and the transport scattering time when going from the ferromagnetic phase to the paramagnetic phase.

PACS numbers: 75.50.Pp, 75.10Lp

The incorporation of Mn atoms into III-V semiconductors by low-temperature MBE techniques was a big advance for the integration of the spin degree of freedom in the semiconductor technology. In Ga$_{1-x}$Mn$_x$As, and for Mn concentrations larger than $x \sim 1\%$, the magnetic ion substitutes a cation, introducing a $S = 5/2$ local moment and a hole in the valence band of the host semiconductor. These compounds present ferromagnetic order, with a Curie temperature, $T_C$, near 100K [1, 2], and this behavior is generally called carrier induced ferromagnetism because the hole carriers mediate the ferromagnetic coupling between the manganese ions [3, 4, 5, 6]. The optimal Mn concentration is near $x \sim 0.05$ and these materials are known as Diluted Magnetic Semiconductors (DMS). Experimentally it is found that the presence of compensating defects reduces the number of carriers in the system being the density of carriers, $p$, much smaller than that of magnetic ions, $c$. For the optimal Mn concentration and not very large carriers densities, most of the magnetic properties of the system seems to be reasonable described by using a virtual crystal approximation (VCA) [3, 4] that neglects thermal and quantum fluctuations as well as disorder effects. These effects are more important as the carrier density in the system becomes larger [3, 5, 6]. Post-growth annealing reduces the number of compensating defects, increases the density of mobile holes in the semiconductor and increases the value of the Curie temperature [7, 8]. In the carrier induced ferromagnetism model $T_C$ is proportional to the carrier density of states at the Fermi energy, and for this reason the annealed samples have a higher $T_C$ than the as-growth samples. Another important difference between the annealed and the as-growth samples is that the later ones present a metal-insulator transition at $T_C$ that is absent in the former ones [10, 11]. In the annealed DMS samples the resistance increases with temperature ($T$) near 25% when going from zero to $T_C$ and remains practically constant when $T$ is further increased. As stated above, the post-growth annealing process reduces the number of defects in the system and the resistivity curves of the annealed samples reflect more defect-free intrinsic properties of the diluted magnetic semiconductor.

Most of the theoretical works on DMS study the origin of the ferromagnetic order: the effects that the electronic properties of the host semiconductor have on the ferromagnetic ground state and the value of $T_C$. In this paper we study the $T$ dependence of the electronic properties of the DMS. We analyze how the spin polarization of the holes affects the electronic properties of the system and from that we obtain the temperature dependence of the dielectric constant and of the conductivity of the doped semiconductor.

The main result of our paper is presented in Fig.1. There, the $T$ dependence of the electrical resistivity for Ga$_{1-x}$Mn$_x$As with $x \sim 0.05$ is plotted for two different hole densities. The variation of the resistivity when going from zero $T$ to $T_C$ is near 20%, in good agreement with experiments in the more intrinsic post growth annealed samples [7, 10]. As we discuss below, this variation of the resistivity with $T$ is due to the dependence of the Fermi surface and scattering times on the carrier spin polarization and therefore on $T$. The model we present explains the observed behavior of the electrical resistivity with temperature.

In our calculations we describe the electronic structure of the DMS using the six band Kohn-Luttinger Hamiltonian and the disorder and thermal effects are treated in a mean field approximation. The dielectric constant is calculated in the Random Phase Approximation (RPA) formalism and the conductivities using the relaxation time approximation.

The system is described by the following Hamiltonian,

$$ H = H_{\text{holes}} + J \sum_{I,I'} S_I \cdot S_I \delta(r_I - r_{I'}) ,$$  

(1)
Where $H_{holes}$ is the part of the Hamiltonian which describes the itinerant holes and the last term is the antiferromagnetic exchange interaction between the spin of the Mn$^{2+}$ ions located at $\mathbf{R}_i$ and the spin $\mathbf{s}_i$ of the carriers. In order to obtain the temperature dependence of the electronic and magnetic properties of the system, we minimize the free energy per unit volume,

$$\mathcal{F} = \mathcal{F}_{ions} + \mathcal{F}_{holes}$$

where $\mathcal{F}_{ions}$ is the contribution of the ion spins to the free energy and in the mean field description has the form,

$$\mathcal{F}_{ions} = -T \log \left( \frac{\sinh \left( \frac{hS}{T} \right)}{\sinh \left( \frac{h}{2T} \right)} \right)$$

being $h = Jp\xi/2$ and $\xi$ the spin polarization of the carriers. $\mathcal{F}_{holes}$ is the free energy of the holes, which is obtained in the virtual crystal approximation (VCA) using a Luttinger $\mathbf{k} \cdot \mathbf{p}$ model for describing the carriers. In the VCA the average density of states for the real system is replaced by that of the average Hamiltonian. This approach implies a translational invariant system with an effective magnetic field acting on the carrier spins $H_{eff} = JS\mathbf{c}$, being $m$ the polarization of the Mn spins. In the Luttinger $\mathbf{k} \cdot \mathbf{p}$ model the wave function of the holes in the state $(n, \mathbf{k})$, where $n$ is the subband index and $\mathbf{k}$ is the wave vector, is expressed as

$$\psi_{n,\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} \sum_{J, m_J} \alpha_{n,\mathbf{k}}^{J, m_J} |J, m_J >$$

where $|J, m_J >$ are the six $\Gamma_{4v}$ valence band wave functions. The coefficients $\alpha_{n,\mathbf{k}}^{J, m_J}$ and the corresponding eigenvalues, $\varepsilon_{n,\mathbf{k}}$, depend on the spin polarization of the Mn, and are obtained, from the Luttinger $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian.\cite{12,13}

By minimizing Eq.(2) we obtain the $T$-dependence of the spin polarization of the carriers, $\xi(T)$ and of the Mn, $m(T)$. In the VCA the Curie temperature has the expression $T_c = 2/3\xi J^2 n_\sigma(\mu)$ where $n_\sigma(\mu)$ is the density of states per site and spin at the Fermi level\cite{12,13}. Through the dependence of the Hamiltonian on the spin polarization, we obtain the temperature dependence of the eigenvalues, eigenvalues and chemical potential. Along this paper we consider always the optimal Mn concentration of $x=0.05$\cite{11} and an exchange coupling $J=0.060eVnm^3$\cite{11}. Although the properties of the system depend on the orientation of the magnetization\cite{2}, this dependence is much smaller than the $T$-dependence and since we are interested in the variation of the electronic properties with $T$ we fix the Mn spin polarization in the $z$-direction.

From the eigenvalues and eigenvectors, we calculate the dielectric constant that in the RPA has the form,

$$\epsilon(q, \omega) = 1 - \frac{4\pi e^2}{\epsilon_0 q^2} \chi(q, \omega)$$

being $\chi(q, \omega)$ the susceptibility,

$$\chi(q, \omega) = \sum_{i,j,k,q} \frac{n_F(\varepsilon_{i,k+q}) - n_F(\varepsilon_{j,k})}{\varepsilon_{i,k+q} - \varepsilon_{j,k} + i\omega} f(i,k)(j,k+q)$$

with

$$f(i,k)(j,k+q) = \left( \sum_{J,m_J} (\alpha_{n,k,i}^{J,m_J} \ast \alpha_{n,k,j'}^{J,m_J})^2 \right)^2$$

where $n_F$ denotes the Fermi-Dirac distribution and $\epsilon_0$ the dielectric constant of the host semiconductor. In the GaAs case $\epsilon_0=12.5$

In Fig. 2 we plot the static susceptibility, $\chi(q, \omega=0)$ as a function of the wave vector for $\mu=0.02nm^3$, and two different temperatures: $T=0$ where the ground state is ferromagnetic with $m=1$ and $\xi=0.777$, and $T=100K> T_c \sim 57K$, where the system is paramagnetic, $m=\xi=0$. The $\mathbf{k} \cdot \mathbf{p}$ Hamiltonian has cubic symmetry, and the susceptibility depends not just on the absolute value of $q$, but also on its orientation. However, for the range of parameters we are interested in, the dependence is almost negligible. We choose always the wavector $q$ pointing to the [100] direction. Furthermore, in the ferromagnetic case, since the magnetic anisotropy is small, the dependence of the susceptibility on the wave vector orientation is very small and not visible in the scale of Fig. 2. The susceptibility depends on $T$ mainly at small wave vectors, and this dependence occurs because the carrier screening ability is proportional to the number of states at the Fermi surface, and this is smaller in the ferromagnetic phase than in the paramagnetic phase. At small wave vectors, only the intraband excitations contribute to the susceptibility and the static dielectric constant can be written as,

$$\lim_{q\to 0} \frac{\epsilon(q)}{\epsilon_0} = 1 + \frac{\lambda_{TF}^2}{q^2}$$

and the T dependence of the screening properties can be characterized by the Thomas Fermi screening length, $\lambda_{TF} \equiv 1/q_{TF}$. In Fig. 3 the $T$ dependence of $\lambda_{TF}$ is illustrated. We also show in the inset of Fig. 3 the $T$-dependence of the ion spin polarization for the same set of parameters. The screening length decreases with $T$ and changes near 8% when going from the zero $T$ ferromagnetic phase to the paramagnetic phase. Since this change is due to the variation of the spin polarization, the Thomas-Fermi length remains constant for $T$ higher than $T_c$.

Finally and in order to compare with experimental information, we analyze the transport properties of the system. In the relaxation-time approximation\cite{17} the conductivity has the form,

$$\sigma_{\alpha,\beta} = \frac{e^2}{hV} \sum_{n,\mathbf{k}} \frac{\tau_{n,\mathbf{k}} \partial \varepsilon_{n,\mathbf{k}}}{\partial k_\alpha} \frac{\partial \varepsilon_{n,\mathbf{k}}}{\partial k_\beta} \left( -\frac{\partial n_F(\varepsilon_{n,\mathbf{k}})}{\partial \varepsilon_{n,\mathbf{k}}} \right)$$
where $\tau_{n,k}$ is the elastic scattering time of the electronic state $(n,k)$, that has the expression,

$$\frac{1}{\tau_{n,k}} = \frac{2\pi}{\hbar} \sum_l C_l Q_l^2 \sum_{n',k'} \left| M_{n,n'}^{k,k'} \right|^2 \times (1 - \cos \theta_{k,k'}) \delta(\varepsilon_{n,k} - \varepsilon_{n',k'})$$

(10)

with

$$M_{n,n'}^{k,k'} = \frac{e^2}{\varepsilon_0 \varepsilon(|k - k'|) |k - k'|^2} \int \rho_{(n,k)}(n',k')$$

(11)

and $C_l$ is the density of scattering defects with charge $Q_l e$. Using equations (10,11) for the scattering time we have assumed that the main scattering centers are Mn$^{2+}$ acceptors, As-antisite defects, and Mn interstitials, all of them have a Coulomb interaction with the carriers. At finite temperature the carriers are also scattered by short-range spin fluctuations[16,17]. However we have neglected the contribution of this spin flip process to the elastic scattering time because the Coulomb contribution is orders of magnitude larger[17,18]. The conductivity of DMS at $T=0$, for different hole densities and different amount of disorder has been calculated recently in Ref. [17]. Here we study the dependence of the conductivity on $T$. We have evaluated expressions [9,10,11] for different spin polarization of the holes, which correspond to different temperatures. Three factors contribute to the variation of the electrical conductivity with $T$. i) A change of the thermal Fermi distribution of the carriers. In the range of temperature of interest this effect produces only a small variation of the electronic properties[10]. ii) The change of $T$ produces a change in the dielectric function Fig.2 and therefore a change in the matrix element Eq.11. However this effect is very weak because in the transport scattering time the forward scattering is suppressed by the $(1 - \cos \theta_{k,k'})$ term and the more important matrix elements have associated a large transfer of momentum for which the electrical susceptibility is practically $T$ independent, see Fig.2. iii) The change from the ferromagnetic to the paramagnetic ground state produces a reduction of the majority spin Fermi surface area. For that reason the wave vector difference that appear in the matrix element Eq.11 is bigger in the ferromagnetic phase than in the paramagnetic phase, and therefore, the scattering time and the conductivity are larger in the ferromagnetic phase than in the paramagnetic phase. This is the main contribution to the variation of the conductivity with $T$.

In Fig.1 we plot the resistivity of Ga$_{1-x}$Mn$_x$As, for $x=0.05$, $J=60meVnm^3$ and two different densities, $p=0.2nm^{-3}$ ($T_C = 57K$) and $p=0.6nm^{-3}$ ($T_C = 125K$). We normalize the resistivity to the value of the resistivity in the paramagnetic phase. As the temperatures studied are always much smaller than the Fermi temperature, in the paramagnetic phase the resistivity is practically $T$ independent. As reported in ref.[13] in the ferromagnetic phase there is an anisotropic effect, and the resistivity is different for directions parallel and perpendicular to the magnetization. The anisotropy is $\sim 2.5\%$ for the $p=0.2nm^{-3}$ case and smaller than $0.5\%$ for the $p=0.6nm^{-3}$ case. More interesting, for both hole densities there is a big change, near $20\%$, in the value of the resistivity when going from zero $T$ to $T_C$. This change is very similar to the experimentally observed[10,11] in the post growth annealed samples. Also the overall shape of the resistivity versus $T$ curve is similar to the experimental one.

Therefore we conclude that our model, that is based in the use of a) the six band $k\cdot p$ model for the band structure of the DMS, b) the mean field approximation for describing the thermal fluctuations, c) the RPA for the dielectric constant and d) the relaxation-time approximation for the resistivity, describe appropriately the electric properties of DMS and their dependence on $T$. Our theory is able to describe adequately the experimental dependence of the resistivity on $T$[10,11].

From our calculations we know that the variation of the resistivity when going from the ferromagnetic phase to the paramagnetic phase is due to the different Fermi surfaces that have these phases. The wave vector transferred in a scattering event across the Fermi surface is larger in the ferromagnetic phase than in the paramagnetic phase and this produces that the resistivity in the paramagnetic phase is larger than in the ferromagnetic phase.

We are grateful to G. Platero for helpful discussions. Financial support is acknowledged from Grants No MAT2002-04429-C03-01 and MAT2002-04095-C02-01 (MCyT, Spain) and Fundación Ramón Areces.
FIG. 2: Static electrical susceptibility as a function of the wave vector for a Mn concentration $x=0.05$, a hole density $p=0.2\text{nm}^{-3}$, an exchange coupling $J=0.06\text{eV nm}^{-3}$ and two temperatures: $T=0$ (Ferromagnetic ground state) and $T=100$K (Paramagnetic ground state). $a$ is the FCC lattice parameter of the host semiconductor.

FIG. 3: Temperature dependence of the Thomas-Fermi length for a Mn concentration $x=0.05$, a hole density $p=0.2\text{nm}^{-3}$, and exchange coupling $J=0.06\text{eV nm}^{-3}$. The inset illustrates the T dependence of the Mn spin polarization.

[1] F. Matsukura, H. Ohno, A. Shen, and Y. Sugawara, Phys. Rev. B 57, R2037 (1998).
[2] H. Ohno, Science 281, 951 (1998).
[3] T. Dietl, H. Ohno, F. Matsukura, J. Cibert, and D. Ferrand, Science 287, 1019 (2000).
[4] T. Jungwirth, W. A. Atkinson, B. H. Lee, and A. H. MacDonald, Phys. Rev. B 59, 9818 (1999).
[5] A. Chattopadhyay, S. D. Sarma, and A. J. Millis, Phys. Rev. Lett. 87, 227202 (2001).
[6] M. J. Caldeiron, G. Gomez-Santos, and L. Brey, Phys. Rev. B 66, 075218 (2002).
[7] H. Ohno and F. Matsukura, Solid State Commun. 117, 179 (2001).
[8] J. Schliemann, J. Konig, and A. H. MacDonald, Phys. Rev. B 64, 165201 (2001).
[9] M. M. Gonzalo Alvarez and E. Dagotto, Phys. Rev. Lett 89, 277202 (2002).
[10] S.J.Potashnik, K.C.Ku, S.H.Chun, J.J.Berry, N.Samarth, and P.Schiffer, Appl. Phys. Lett. 79, 1495 (2001).
[11] K. W. Edmonds, K. Y. Wang, R. P. Campion, A. C. Neumann, N. R. S. Farley, B. L. Gallagher, and C. T.
Foxon, Appl. Phys. Lett. 81, 4991 (2002).
[12] T. Dietl, H. Ohno, and F. Matsukura, Phys. Rev. B 63, 195205 (2001).
[13] J. B. M. Albofath, T. Jungwirth and A. H. MacDonald, Phys. Rev. B 63, 054418 (2001).
[14] J. Okabayashi, A. Kimura, O. Rader, T. Mizokawa, A. Fujimori, T. Hayashi, and M. Tanaka, Phys.Rev.B 58, R4211 (1998).
[15] T. Jungwirth, M. Abolfath, J. Sinova, J. Kucera, and A. H. MacDonald, cond-mat/0206416.

[16] M. Fisher and J.S.Langer, Phys. Rev. Lett. 20, 665 (1968).
[17] F.C.Zumsteg and R.D.Parks, Phys. Rev. Lett 24, 520 (1970).
[18] M. J. Calderón, J.A.Vergés, and L. Brey, Phys. Rev. B 59, 4170 (1998).
[19] Note that the Fermi temperature is much higher than any other temperature in the system. In particular than the Curie temperature.