Optical conductivity of Mn-doped GaAs

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Abstract. The optical conductivity of the III–V diluted magnetic semiconductor compound (Ga, Mn)As was investigated theoretically. A direct comparison between the calculations and available experimental data is provided. We demonstrate that our model study is able to reproduce both qualitatively and quantitatively the measurements carried out by Burch et al (2006 Phys. Rev. Lett. 97 087208) and Singley et al (2002 Phys. Rev. Lett. 89 097203, 2003 Phys. Rev. B 68 165204). It is found that an increase of the carrier density by up to one hole per Mn leads to a redshift of the broad conductivity peak located at approximately 200 meV in optimally annealed samples. Our study demonstrates that the non-perturbative treatment (beyond the valence band picture) is crucial for capturing these features. Otherwise a blueshift and an incorrect amplitude would result. We have calculated the Drude weight (order parameter) and have established the metal–insulator phase diagram. It is shown that (i) Mn-doped GaAs is indeed close to the metal–insulator transition and (ii) in both 5 and 7% doped samples, only 20% of the carriers are delocalized. Beyond the mobility edge, we found that the optical mass is $m_{\text{opt}} \approx 2m_e$. Interesting new features of overdoped samples were found. The overdoped regime could be experimentally realized by Zn codoping. Detailed discussions and a careful analysis are provided.

The possibility of using for spintronics purposes the spin degree of freedom in magnetic semiconductors has led to tremendous experimental and theoretical work. One of the main goals is to search for optimal candidates that exhibit high Curie temperatures (beyond room temperature). Although it is well accepted that the presence of holes in III–V diluted magnetic

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semiconductors is responsible for the induced ferromagnetism, the particular case of (Ga, Mn)As remains controversial concerning the nature of the states at the Fermi level. There are, in fact, several important points that are still controversial in the literature. Is the Fermi level inside the weakly perturbed valence band (VB scenario) or does (Ga, Mn)As exhibit a well-defined impurity band? Are the holes extended or localized? Or is the situation similar to that of Mn-doped II–VI as (Zn, Mn)Te? For those latter compounds the VB picture is appropriate. It seems that this could be at the origin of some of the controversies. In our view the main question should be: In the case of (Ga, Mn)As, is a perturbative treatment (VB picture) appropriate to explain the physics? Or equivalently, is it necessary to treat disorder effects properly? From ab-initio-based studies, one finds that (Ga, Mn)As clearly exhibits a preformed impurity band [1]–[3] that is responsible for (i) the non-Ruderman–Kittel–Kasuya–Yosida (RKKY) nature of the magnetic couplings and (ii) explains the measured high Curie temperatures (with respect to II–VI compounds). Additionally, it has been clearly demonstrated that RKKY couplings obtained within the VB picture cannot explain the magnetism in III–V compounds [4]. Indeed, the frustration effects would lead either to a spin glass phase or to extremely small Curie temperatures. On the other hand, the non-perturbative treatment of the appropriate model Hamiltonian captures the essential physics and leads to non-RKKY couplings [5], in agreement with ab initio studies. Note also that below 1% the impurity band is completely split from the VB. Early resistivity measurements have clearly demonstrated that (Ga, Mn)As is close to the metal–insulator phase transition. Indeed, measurements in as-grown samples often show insulator behavior at low temperature and metallic after annealing [6]–[9]. Of course, the VB scenario is unable to capture this physics, namely the metal–insulator transition. It has been shown by Singley et al [10, 11] and Burch et al [12] that the measured optical conductivity is also inconsistent with the perturbative VB scenario. The aim of this paper is to provide a non-perturbative theoretical study able to reproduce and explain the experimental observations.

Ab-initio-based studies have provided the most reliable tool, to allow for quantitative studies of the magnetic properties without any adjustable parameters. As an example, it was possible to study in great detail and quantitatively the magnetic properties of Ga$_{1-x}$Mn$_x$As, both in the presence and absence of native (compensating) defects [13]–[15]. However, a dynamical transport study is not a simple task within the ab-initio approach. A study of the relevant minimal model is thus necessary. The model approach is especially suitable for understanding the influence of a particular physical parameter and may provide support for finding new potential spintronic candidates. The V–J model treated non-perturbatively was shown to capture qualitatively the magnetic properties of a whole family of III–V materials [5]. The present theoretical study is based on such a model. The one-band V–J Hamiltonian reads

$$H = -\sum_{ij} t_{ij} c_i^{\dagger} c_j + \sum_i p_i J_i \mathbf{S}_i \cdot \mathbf{s}_i + \sum_{i\sigma} \epsilon_i p_i c_i^{\dagger} c_i \cdot \sigma .$$

In the first term, $t_{ij} = t$ if $i$ and $j$ are nearest neighbors, otherwise $t_{ij} = 0$. The random variable $p_i = 1$ if the site is occupied by an impurity, otherwise it is 0. $\mathbf{s}_i$ denotes the localized impurity spin at site $i$ ($|\mathbf{s}_i| = 5/2$) and $\mathbf{S}_i$ denotes the spin-1/2 operator of the carrier. $J_i$ is the $p$–$d$ coupling ($J$) between the itinerant carrier ($p$-states) and the localized Mn spin. The last term results from the substitution of Ga$^{3+}$ by Mn$^{2+}$: $\epsilon_i = V$ if the site is occupied by Mn, otherwise it is 0. This additional crucial term allows us to adjust the position of the hybridized $p$–$d$ states with respect to the top of the VB [5, 16]. In the following, $x$ denotes the Mn concentration and $p$ the hole density. In the absence of compensating defects (As anti-sites or Mn interstitials)
The calculations are performed by exact diagonalization of the Hamiltonian (1) in both spin sectors. This provides the set of eigenvalues and eigenstates \( \{ E^{\sigma, c}_{\tau}, |\Psi^{\sigma, c}_{\tau}\rangle\} \), where \( \sigma = \uparrow, \downarrow \) and \( r = 1, 2, \ldots, N \), that are used to evaluate the optical conductivity \( \sigma(\omega) \) and the Mn–Mn magnetic couplings. The superscript \( c \) denotes the configuration of disorder; it will be omitted in the following. Note that the calculations are performed at \( T = 0 \) K assuming that the impurity spins are aligned.

For simplicity, our calculations are performed on a simple cubic lattice instead of the fcc lattice of Ga in the real GaAs compound. Thus, instead of 4 atoms per unit cell we have only 1. Hence, the value of the lattice spacing that will be used is \( a = (a_0/4^{1/3}) = 3.55 \times 10^{-10} \) m, where \( a_0 = 5.65 \times 10^{-10} \) m is the lattice parameter of the zinc-blende GaAs material. Let us briefly justify this choice. In these diluted compounds the carrier densities are very low; thus it is natural to expect the structure of the lattice to play no relevant role. As will be seen, this simplification will not have a significant effect on the results. This is also valid when the carrier is localized, as far as the localization length remains much larger than the lattice spacing.

Let us now discuss how the model parameters are fixed. First, the hopping integral \( t \) is chosen according to the bandwidth \( W \) of the GaAs host. Thus, we use \( t \approx 0.7 \) eV \( (W = 12t) \). \( JS \) is set to 3 eV since it is now well accepted that in \((\text{Ga, Mn})\text{As}, J \approx 1.2\) eV \([17, 18]\). Note that \( J \) is also of the order of 1 eV for other materials including II–VI compounds \((\text{Zn, Mn})\text{Te}, (\text{Cd, Mn})\text{Te}, \text{etc})\) \([19]–[23]\). The last parameter \( V \) (on-site energy) is, as mentioned above, the crucial one. It actually explains the different nature of the Mn–Mn couplings and Curie temperatures values in Mn-doped compounds such as \((\text{Zn, Mn})\text{Te}, (\text{Ga, Mn})\text{As} \) and \((\text{Ga, Mn})\text{N})\). The on-site energy \( V \) contains two contributions: (i) an electrostatic one due to the difference of charge between the host and substituted cation and (ii) another resulting from the Schrieffer–Wolff \([24]\) transformation of the Anderson Hamiltonian. Indeed, in addition to the \( \sum_i p_i S_i \cdot s_i \) term, this transformation leads to a spin-independent scattering contribution that depends on the position of the \( d \)-levels with respect to the top of the VB in the host material. \( V \) is set in order to reproduce the energy of the acceptor level \( E_a = 110 \) meV in the GaAs host \([25]–[28]\). This leads to \( V = 1.25 \) eV = 1.8\( t \). Note that with this set of parameters, we were able to reproduce the whole variation of the Zeeman splitting as a function of \( x \) obtained within first principle studies \([29, 30]\). This splitting is defined by \( \Delta(x, V) = (1)_{\text{max}} - (1)_{\text{max}} \), where \( (1)_{\text{max}} \) is the largest eigenvalue in the \( \sigma \)-sector \([16]\). For example, from \textit{ab-initio} calculations \([29, 30]\), \( \Delta_{\text{ab-initio}} = 0.65 \) eV for \( x = 0.05 \), we obtain a value of 0.66 eV. Thus, from now on, the set of three parameters \((t, J, V)\) is fixed, and the calculations that will follow are performed without any adjustable additional parameter. The calculations are performed for both 5 and 7% doped compounds in the ferromagnetic phase. Experimental and theoretical studies have shown that below 1% no long-range ferromagnetic order is possible (below the percolation threshold). In the very dilute regime (below 1%), the optical conductivity was recently studied in \([31]\) within a very different theoretical approach. They perform some variational calculations on the Mn–Mn molecule and extract the model parameters that are used to perform their calculations.
Let us now explain in more detail our theoretical method. The total optical conductivity reads $\sigma(\omega) = \sum_{\sigma} \sigma_{\sigma}(\omega)$, where $\sigma_{\sigma}(\omega)$ is given by the following expression [32]–[34]:

$$\sigma_{\sigma}(\omega) = D_{\sigma} \delta(\omega) + \sigma_{\sigma}^{\text{reg}}(\omega).$$  \hspace{1cm} (2)

$D_{\sigma}$ is the Drude weight ($\omega = 0$ contribution) in the $\sigma$-sector and $\sigma_{\sigma}^{\text{reg}}(\omega)$ is the regular part of the optical conductivity. Note that the Drude weight is the order parameter for the metal–insulator phase transition: it is zero in the insulating phase and finite in the metallic one. In our calculations, we have used periodic boundary conditions in order to separate the finite frequency contribution from the dc part. In that case, the Kubo expression provides $\sigma_{\sigma}^{\text{reg}}(\omega)$ only. The Drude weight $D$ is then obtained from the following sum-rule:

$$D_{\sigma} = -\int_{0}^{\infty} \sigma_{\sigma}^{\text{reg}}(\omega) d\omega - \frac{\sigma_{0}}{\hbar} \left\langle \hat{K}_{x}^{z} \right\rangle / N.$$  \hspace{1cm} (3)

$\left\langle \hat{K}_{x}^{z} \right\rangle$ is the average hole kinetic energy in the $x$-direction and $\sigma$-sector, $N = L^{3}$ is the total number of sites and $\sigma_{0} = (\pi e^{2}/\hbar a) = 21540 \Omega^{-1}\text{cm}^{-1}$.

To allow for a direct comparison between available experimental data and theory, we have to take into account the following point: the substitution of $\text{Ga}^{3+}$ by $\text{Mn}^{2+}$ provides $n_{i} = 3$ $p$–$d$ states near the top of the VB (see figure 1 for an illustration). These states are known as the dangling bond hybrid states [35]. As mentioned before, in the case of a single Mn impurity, the acceptor level energy is $E_{b} \approx 0.11 \text{eV}$. At finite concentration, an impurity band is formed and merges into the VB when $x$ becomes large enough, $x \geqslant 0.010$. Because $\text{Mn}^{2+}$ provides 1 hole per Mn, the resulting $p$–$d$-hybridized states impurity band is thus one-third filled. On the other hand, in the one-band $V$–$J$ model, each impurity provides a single state. Thus, to be consistent and coherent, if the (Ga, Mn)As compound contains a concentration $x$ of Mn and $p$ of holes, the theoretical calculations should be performed as follows: (i) the one-band calculations are performed for the hole density $\tilde{p} = p/n_{i}$ and (ii) the theoretical optical conductivity that is compared to the experimental data is

$$\sigma(\omega, p) = n_{i} \tilde{\sigma}(\omega, p/n_{i}),$$  \hspace{1cm} (4)

where $\tilde{\sigma}(\omega, p/n_{i})$ is that calculated within our one-band model. Similarly, the Curie temperature will be performed using the Mn–Mn magnetic couplings,

$$J_{i,j}(x, p) = n_{i} \tilde{J}_{i,j}(x, \tilde{p} = p/n_{i}),$$  \hspace{1cm} (5)
Figure 2. Illustration of the procedure for calculation of the magnetic couplings between two Mn impurities located at sites $i$ and $j$. The left part corresponds to the real compound in which each Mn$^{2+}$ provides three $p$–$d$-hybridized states and one hole (this picture describes three Mn impurities in GaAs). The right side corresponds to the one-band $V$–$J$ model. The $c_{ij}^{r,r'}$ are the matrix elements that enter the expression for the exchange given in equation (7).

where $\tilde{J}_{i,j}(x, p/n_l)$ are those calculated within the one-band $V$–$J$ model. To be more specific, these couplings are given by the following generalized susceptibility,

$$\tilde{J}_{i,j}(x, p) = -\frac{1}{4\pi S^2} \int_{-\infty}^{E_F} \text{Tr}(\Sigma_i G_{i,j}^{\uparrow}(\omega) \Sigma_j G_{j,i}^{\downarrow}(\omega)) \, d\omega,$$

where the one-particle retarded Green’s functions $G_{i,j}^{\sigma}(\omega) = \langle i\sigma | 1/(\omega - \hat{H} + i\epsilon) | j\sigma \rangle$. In the present, the local exchange splitting energy is a constant $\Sigma_i = JpdS$. An illustration of the procedure that provides the Mn–Mn exchanges is shown in figure 2. Note that this picture also applies for the optical conductivity calculation. In this case, the matrix elements $c_{ij}^{r,r'}$ are replaced by those involving the current operator (see below). The previous equation can be rewritten as follows,

$$\tilde{J}_{i,j}(x, p) = -\frac{1}{4S^2} \sum_{r,r'} c_{ij}^{r,r'} n_{r}^{\uparrow} - n_{r'}^{\downarrow},$$

where $n_{r}^{\sigma}$ is the occupation number associated with the state $|\Psi_{r}^{\sigma}\rangle$. The coefficients $c_{ij}^{r,r'} = A_{ij,r}^{\uparrow} \cdot A_{ji,r'}^{\downarrow}$, where $A_{ij,r}^{\sigma} = \langle i | \Psi_{r}^{\sigma} | j \rangle$. The matrix elements $c_{ij}^{r,r'}$ contain the relevant information about the nature of the hole states and thus the nature of the magnetic couplings. In the VB picture, $r$ (respectively $r'$) are replaced by the momentum $\vec{k}$ (respectively $\vec{k}'$) and $c_{ij}^{r,r'} = (1/N) e^{i(\vec{k} - \vec{k}' ) \cdot (r - r')} \text{leading to the standard RKKY couplings. Diagonalization of the resulting effective dilute Heisenberg Hamiltonian} \, \hat{H}_{\text{Heis}} = \sum_{ij} p_i p_j J_{i,j}(x, p) S_i \cdot S_j \, \text{will provide the Curie temperature} \, T_C(x, p). \, \text{This procedure will be done within the self-consistent local random-phase approximation (RPA) (SC-LRPA) [14]. The SC-LRPA is a semi-analytical Green’s function-based method that is in particular able to capture quantitatively both the physics of localization and percolation. The disorder is treated in the real space (no effective medium) and the thermal/transverse fluctuations beyond standard mean field within the RPA. Comparisons with Monte Carlo treatment have demonstrated several times that the SC-LRPA is a reliable and very accurate tool (see for example [14, 36]).}
Figure 3. Regular part of the optical conductivity $\sigma(\omega)$ as a function of $\omega/t$ for $x = 0.05$: (left) for ‘underdoped’ samples (electron-doped), e.g. from low carrier concentration to $p/x = 1$ (well-annealed sample), (right) ‘overdoped’ samples (hole-doped). The parameters are $JS = 4.3t$ and $V = 1.8t$ ((Ga, Mn)As set). The optical conductivity is given in units of $\sigma_0$ (see text).

The regular part of the optical conductivity $\sigma_{\sigma}^{\text{reg}}(\omega, p/n_i)$ is given by the Kubo expression, which reads

$$
\sigma_{\sigma}^{\text{reg}}(\omega, p/n_i) = \frac{\sigma_0}{N} \sum_{r \neq r'} (n_{r}^{\sigma} - n_{r'}^{\sigma}) \frac{A_{rr'}^{\sigma}}{E_{r'}^{\sigma} - E_{r}^{\sigma}} \delta(h\omega - E_{r'}^{\sigma} + E_{r}^{\sigma}).
$$

(8)

The matrix element $A_{rr'}^{\sigma} = |\langle \Psi_f^{\sigma} | \hat{j}_x | \Psi_i^{\sigma} \rangle|^2$, where $\hat{j}_x = -i \sum_{ij} (c_{i,j,\sigma}^\dagger c_{i+\hat{x},\sigma} - h.c.)$ is the $x$ component of the current operator in the $\sigma$-sector.

In figure 3, we have plotted, for $x = 0.05$ and various carrier densities, the regular part of the optical conductivity as a function of frequency. The calculations are realized on a $20^3$ site system. A comparison with those performed on both $16^3$ and $24^3$ systems has shown that the finite size effects are negligible. A systematic average over at least 200 configurations of disorder has been done. As observed experimentally, $\sigma(\omega)$ exhibits a clear broad peak for all carrier concentrations. As the hole density increases from the lowest concentration up to optimal doping $p/x = 1$ (left panel of figure 3), we observe a clear redshift of the peak position. This feature is in agreement with the detailed experimental study performed by Burch et al (see the left panel of figure 3 in [12]). Moreover, this is in contrast to the VB picture, which predicts a blueshift [37] (see also the right panel of figure 3 in [12]). Note that a blueshift was reported in recent measurements [38] (see figure 1(c) in the preprint [38]), but no clear explanation was given to explain the origin of the conflict between earlier studies and their recent results. The data shown in figure 1(c) in [38] result from a fit procedure that depends on many parameters. Thus, it would be of great interest and very useful if the authors could show the measured data; this would facilitate the comparison with earlier studies. On the other hand, we have noticed
that the annealed samples appear to be well annealed. Indeed, the measured Curie temperatures $T_C$ (shown in table I of [38]) agree very well with (i) the first principle-based calculations [14] and with those obtained in the present study (see below).

From the left panel of figure 3, we observe that the peak is located at $0.2t$ for the uncompensated sample ($p = x$) and approximately 0.35$t$ for the lowest concentration, which corresponds to the presence of about 25% of compensating native defects as As antisites (double donors of electrons) for example. A direct quantitative comparison with the experimental data will be given in the next section. To our knowledge, no experimental data are available for larger hole density (beyond 1 hole per Mn); nonetheless, we have also analyzed the effect of co-doping (Ga, Mn)As with a hole donor. This can be achieved experimentally by the substitution of Ga$^{3+}$ by Zn$^{2+}$ for example (Zn$^{2+}$ introduces 1 hole per Zn). We observe in the right panel of figure 3 that adding holes leads to an increase of the conductivity and now to a blueshift of the peak to much higher energy. For example, for $p/x \geq 2.1$, the peak is now located at approximately 1.0$t$. Such a hole concentration could be obtained in Ga$_{0.9}$Mn$_{0.05}$Zn$_{0.05}$As for example. It would be interesting to analyze experimentally the variation of $\sigma(\omega)$ as a function of $y$ in the series of compounds Ga$_{0.95-y}$Mn$_{0.05}$Zn$_{y}$As after annealing. Let us now discuss in more detail the origin of both the redshift and blueshift obtained. For different hole density concentrations, we have been able to isolate the processes that dominate in the optical conductivity response. In other words, we have identified which matrix elements $A_{\nu\nu'}^\tau$ are responsible for the peak in $\sigma^{\rm reg}(\omega, p)$. We could identify three relevant regions denoted (1), (2) and (3) depicted in figure 4. The origin of the redshift is explained in cartoon (a): below $p/x = 1$, the only possible transitions are between regions (1) and (2) (transitions are only allowed between occupied and empty states). The transitions between (1) and (3) are much smaller. As the hole density increases up to $p/x = 1$, the Fermi energy moves down and the energy $\omega_{12}$ associated with $A_{12}$ processes decreases from 0.4$t$ to 0.2$t$. This is an explanation for the redshift. In the very dilute limit (typically below 1%), this corresponds to the process between the well-defined (split) impurity band and the top of the VB as discussed in [12].

For $1 \leq p/x \leq 2$ (case (b)) the Fermi level lies now in the region (2). The two relevant matrix elements are of type $A_{12}$ and $A_{23}$. As the hole density increases, the relative weight of $A_{23}$ increases, while $A_{12}$ decreases. This can be seen in the right panel of figure 3 for $p/x = 1.35$ and 1.95, where a shoulder coming from the $A_{23}$ processes is clearly visible and becomes larger as the hole density increases. The typical energy associated with $A_{23}$ processes is of the order of $\omega_{23} \approx t$. As we further increase the hole density (case (c)), $A_{12}$ processes are now forbidden (regions (1) and (2) are empty or filled with holes) and the only possible transitions are between regions (2) and (3). Thus, this explains the peak located at $\omega/t \approx 1$ for $p/x \geq 2$ and the origin of the blueshift.

We now proceed further and discuss the variation of the metal–insulator order parameter, the Drude weight, as a function of the carrier concentration. Note that the dc conductivity can be obtained from $\sigma_{\text{dc}} = D\tau$, where $\tau = l_e/\nu_F$ with $l_e$ being the mean free path. In the top panel of figure 5, the averaged Drude weight $D_L$ is plotted as a function of the carrier density per Mn for various system sizes. The Mn concentration is set to $x = 0.05$ and $L$ varies from $L = 12$ to 20. For the smallest system, an average over 1000 configurations of disorder has been performed, and for the largest, 200 configurations were used. After performing an extrapolation in the thermodynamic limit (see the bottom panel of figure 5), we obtained the metal–insulator phase diagram plotted in the same figure. As seen, below a critical concentration (mobility edge) the Drude weight $D$ is zero: the system is an insulator. Above this threshold it scales linearly with
Figure 4. The top of the figure is a sketch of the density of states for Mn-doped GaAs: a compound that contains typically 5% Mn. A zoom of the preformed impurity band is also shown. There are three relevant regions that provide the dominant transition matrix elements in the Kubo formula of the optical conductivity (see equation (8)) as described below. There are three relevant regions. Region (1) corresponds to the localized states (the mobility edge $E_C$ is indicated) and both regions (2) and (3) correspond to disordered extended states. (a) For $p/x \leq 1$ the dominant transition matrix element is $A_{12}$ between regions (2) and (1); (b) for $1 \leq p/x \leq 2$, the matrix elements $A_{12}$ decrease and $A_{23}$ (between regions (2) and (3)) are now allowed and start to dominate; (c) for $2 \leq p/x \leq 3$, $A_{12}$ contributions are not possible anymore and the dominating process is $A_{23}$. Note that the width of the vertical arrows indicates the weight of the processes.

the delocalized (extended) hole concentration, $p_{ex} = (p - p_c)$. The transition point is located at $(p/x)_c \approx 0.80$ for both Mn concentrations $x = 5$ and $x = 7.3\%$. Thus, our present study predicts that the critical carrier density is $p_c \approx 0.0375$ for the 5% doped sample and $p_c \approx 0.055$ for $x = 0.073$. This result is important since it indicates that only approximately 20% of the carriers contribute to the Drude weight, while most of them (80%) are localized. Beyond the critical concentration, the slope of the Drude weight $D$ provides the value of the optical masses (for localized carriers $m_{opt} = \infty$). Indeed, in this regime, one can write $D(p, x)/\pi = p_{ex}e^2/m_{opt}$. We find that $m_{opt}(x = 0.073) \approx m_{opt}(x = 0.05) \approx 4m_0 = 2/t$, where $m_0 \approx 0.5m_e$ is the hole effective mass in the GaAs host. All these results are consistent with the experimental observation that (Ga, Mn)As is close to the metal–insulator phase transition and why as grown insulating samples become metallic after annealing. Note that from their experimental data, Singley et al [10] have estimated $m_{opt}$ to be within the range $0.7m_e \leq m_{opt} \leq 15m_e$. New Journal of Physics 13 (2011) 023002 (http://www.njp.org/)
The natural question that arises now is the following: Is our theoretical model also able to explain the experimental data quantitatively? Let us then compare directly our calculations with the experimental data of the optical conductivity measured in annealed samples [12]. We will see that our model theory is able to reproduce the optical conductivity data and the Curie temperatures simultaneously. In figure 6, we have plotted the optical conductivity in absolute units (Ω cm$^{-1}$) as a function of the energy (in eV). The experimental data for the annealed samples are taken from [12]. The calculations were performed assuming 1 hole per Mn (perfectly annealed samples), taking into account, as discussed before, the fact that each Mn impurity brings three hybridized $p$–$d$ states. We observe very good and surprising quantitative agreement for both concentrations of Mn. It is even excellent in the case of the 5% doped sample. The peak position is located at 200 meV for $x = 0.05$ in perfect accordance with the measurements,
and the overall shape (peak structure, width) is also very well reproduced. Furthermore, using the calculated couplings $J_{ij}(x, p)$ in the effective diluted Heisenberg Hamiltonian (see above), we have calculated within SC-LRPA the Curie temperature for both cases. We have found that $T_C$ is, respectively, 120 and 150 K for $x = 0.05$ and 0.07. These values agree very well with those measured in these annealed compounds, namely 120 and 140 K, respectively [12]. Note also that our calculated values agree very well with those obtained using the ab-initio couplings [14]. It is interesting to remark that if instead of $a = a_0/\sqrt{3}$ we had used $a_0$, then the calculated optical conductivity would have been 60% smaller. The small deviation between theory and experiment observed in figure 6 for $x = 0.073$ may be explained as follows. We have used, for the comparison in both cases, the total measured optical conductivity (the Drude part included). As seen in figure 1 of [12], a small shoulder is seen at 30 meV. The Drude contribution is larger in the $x = 0.073$ compound than in the 5% doped one and this may thus explain why, below the peak, the measured optical response is a bit larger than the calculated one.

To conclude, we have calculated in a non-perturbative way the optical conductivity, the phase diagram and the Drude weight as a function of the hole density in GaAs samples doped with 5 and 7.3% of Mn. In agreement with experimental measurements [12], we have obtained a redshift of the optical conductivity peak. We were also able to reproduce quantitatively the measured optical conductivity in well-annealed samples. Additionally, the calculated Curie temperatures are in excellent agreement with those obtained using ab-initio couplings and with the experimental values. This success shows that our minimal model is able to capture not only qualitatively but also quantitatively both transport and magnetic properties of diluted magnetic semiconductors.

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Figure 6. Calculated and measured optical conductivity $\sigma(\omega)$ in Ga$_{1-x}$Mn$_x$As (in units of $\Omega^{-1}$ cm$^{-1}$) as a function of $\omega/t$ for the optimal doping (1 hole per Mn) and for both Mn concentrations $x = 0.05$ and 0.073.
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