Optics and magnetism
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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2004

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Summary

In this thesis, optical spectroscopy has been used to get an insight in the physical properties of various strongly correlated systems. In general, optical spectroscopy is an excellent tool to study the electronic structure of a given material because its complex optical conductivity gives information about how the electrons move inside the material in response to an external field. A particular process described by the optical conductivity is the motion of the electrons from one site to the other. This process is determined by the competition between electron-electron correlations (which tends to localize the electrons) and wave function hybridization (which tends to delocalize them). It is precisely this competition which is the essence of strong correlations.

Itinerant magnetism: Silicides

The first system described in this thesis contains a transition metal and silicon. In particular the silicides containing manganese, iron and cobalt were studied. Although these transition metals present magnetism in its elemental form, the corresponding silicides, surprisingly, are not magnetic at room temperature. In fact, at low temperatures, the only one that is magnetic is MnSi ($T_C = 30$ K). Above $T_C$ it follows a Curie-Weiss law with a magnetization much larger than the saturation magnetization in the magnetic phase. This define MnSi as a weak ferromagnet. Weak ferromagnetism also appears in a large region of the solid solutions between FeSi and CoSi despite the fact both of them are not magnetic. In contrast, starting from MnSi, replacing Mn by Fe destroys rapidly the weak ferromagnetism. Some of the magnetic solutions of Fe$_{1-x}$Co$_x$Si were also studied in this thesis.

In these silicides, all the stoichiometric compounds and their solutions are bad metals at room temperature. When the temperature is decreased, however, the behaviour is diverse. The resistivity of MnSi decreases strongly and remains metallic down to the lowest temperature without showing superconductivity. Replacing Mn by Fe, although decreases $T_C$, only decreases the rate at which the resistivity decreases with temperature. Surprisingly, in a thin sleeve around FeSi (towards both MnSi and CoSi), the system shows an insulating ground state the
origin of which is still matter of debate. Towards CoSi and beyond this insulating sleeve, the system remains metallic with a small decrease of the resistivity. The behaviour of the resistivity in magnetic fields is also different between the magnetic samples around MnSi and those in Fe$_{1-x}$Co$_x$Si. Around MnSi, the system shows negative magnetoresistance as usually expected for a metal at not so low temperatures. In the Fe$_{1-x}$Co$_x$Si samples the opposite has been found and the detailed behaviour is even analogous to the quantum effects induced by disorder at extremely low temperatures.

Weak ferromagnetism is usually explained in an itinerant picture which was briefly described in Chapter 1. Although it explains various properties of weak ferromagnets, it fails in others. Moreover, it does not provide an explanation of the different behaviours presented in the weak ferromagnets around MnSi and those in Fe$_{1-x}$Co$_x$Si. In this thesis (Chapter 3) we addressed several of these failures regarding the transport and optical properties of MnSi. We showed that the optical properties of MnSi at low temperatures can be described by a phenomenological formula that contains the Drude description as a particular case. The implication is that the temperature dependence of the scattering rate (extracted from the DC resistivity) and its frequency dependence (extracted from the optical conductivity) do not follow the same dependence. This has enormous implications since temperature and frequency are only different manifestations of energy.

In Chapter 4 of this thesis FeSi, CoSi and various of its solid solutions were studied. It was shown that the formation of the insulating state in FeSi is not conventional in the sense that it is not fully explained by band theory. Two observations were determinant for this conclusion. First, the gap of the insulating state is filled at temperatures much lower than would be permitted by thermal excitations only. Second, it was demonstrated that the optical spectral weight contained in the optical conductivity (i.e. the integrated area below it) is not recovered in an energy range much larger than the insulating gap. This lack of recovery points to the necessity of taking into account strong correlations to describe the insulating phase of FeSi. Moreover, strong correlations would explain the magnetotransport properties of Fe$_{1-x}$Co$_x$Si. At relatively high temperatures, these properties are similar to the ones shown by strongly disordered metals at sub-kelvin temperatures.

**Local Moments and Itinerant Electrons: Heavy Fermion Systems**

Several systems show clearly two kinds of electrons. On one hand there are itinerant electrons that are responsible for the transport properties. On the other hand there are electrons that are localized and are responsible of the magnetic properties. When these electrons interact it gives rise to a rich behaviour. One of such systems is composed by the so called heavy fermion materials. This name
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originate from the fact that their properties are similar to those of the normal metals, though with much larger proportional constants. Since in the usual theory of metals these constants are proportional to the electron mass, the enhanced properties of heavy fermion systems can be interpreted as a renormalized mass of the electrons due to strong interactions.

Chapter 5 of this thesis studies a particular family of heavy fermion systems: CeMIn$_5$ (M = Co, Rh, Ir). This system has recently attracted a considerable attention due to the variety shown in its phase diagram. As the temperature is decreased, heavy fermion behaviour appears and precedes states like antiferromagnetism in CeRhIn$_5$ ($T_N = 3.8$ K) and unconventional superconductivity in CeCoIn$_5$ ($T_c = 2.4$ K) and CeIrIn$_5$ ($T_c = 0.8$ K). It has also been suggested that the superconductivity in these materials is similar to the high $T_c$ superconductors. The analogy even starts in the crystal structure that can be seen as layers of CeIn$_3$ (a well known heavy fermion system) separated by MIn$_2$ layers.

The work presented in this thesis has concentrated on the optical properties of the heavy fermion state. It has been shown that, at low temperatures, the optical responses of CeCoIn$_5$ and CeIrIn$_5$ are characterized by a narrow Drude peak representing the oscillations of the coherent electrons. This peak is formed below a certain characteristic energy known as hybridization gap. On the other hand, antiferromagnetism competes against the formation of the coherent state. This is shown in CeRhIn$_5$ where the narrow Drude peak does not evolve completely. Moreover, from the study of its dielectric function it was shown that the hybridization gap is smaller than the other two compounds.

Local Moments and Itinerant Electrons: Manganites

Finally, materials corresponding to the family of perovskite manganites were studied. Perovskite manganites are well known for having a phase diagram with multifarious phases. An example of this is the La$_{1-x}$Ca$_x$MnO$_3$ system. At low temperatures the phase is canted antiferromagnetic in LaMnO$_3$ and CaMnO$_3$ but between them various phases are found: ferromagnetic insulating and metallic, charge ordering, and antiferromagnetism. In general terms, this rich behaviour is the result of the interaction between localized moments and itinerant electrons in the manganese site. At least in the ferromagnetic metallic, this is well described by the double exchange Hamiltonian. However, other ideas and mechanisms have to be considered for a complete description of the manganites. One of them is strong electron-phonon interactions as is revealed in several forms, for example, the large isotope effect. Another mechanism that has to be considered is the natural tendency of the double exchange Hamiltonian to produce phase separation.

The final Chapter of this thesis describes the optical response of thin films of (La$_{0.5}$Pr$_{0.5}$)$_{0.7}$Ca$_{0.3}$MnO$_3$ grown in SrTiO$_3$ substrates. Samples containing different oxygen isotopes, $^{16}$O and $^{18}$O, were studied. At room temperature,
both kind of films show identical DC resistivity with no long range magnetic order. When the temperature is lowered, the resistivity increases until a certain temperature, $T_C$, from which it decreases again. This critical temperature is also accompanied by magnetic ordering. Samples with both isotopes show the same qualitative behaviour just described. However, samples containing $^{18}$O show a lower $T_C$ and the resistivity well below $T_C$ is one order of magnitude larger than its counterpart. Another difference is evident when the temperature is raised above $T_C$. Samples with $^{18}$O show a large hysteresis while in samples with $^{16}$O, it is small. It has been argued that this is a manifestation of phase separation.

As in the DC resistivity measurements, the optical responses of both films are identical at room temperature but greatly differ at low temperatures and especially at low frequencies. At high frequencies (above 2.5 eV), the optical conductivity of both films is dominated by transitions corresponding to charge transfer excitations that are weakly temperature dependent. At low frequencies, above their corresponding $T_C$’s, the optical conductivity of both films is dominated by a strong mid-infrared peak at around 1.5 eV (peak 3). This peak is favored in the paramagnetic state as it corresponds to intersite transitions $e_g^1(Mn^{+3}) \rightarrow e_g(Mn^{+4})$ where the transferred electron ends up being antiparallel to the localized electrons in the $t_{2g}$ level. The intensity of peak 3 remains more or less constant above $T_C$ but decreases rapidly below it. However, it does not disappear completely at low temperatures, particularly in the film containing $^{18}$O where remains rather large. Another peak, labeled 2, can be identified at even lower frequencies (0.75 eV). This peak was identified also as an intersite transition of the same types as peak 3 with the difference that the transferred electron is parallel to the $t_{2g}$ core. In this way, it is enhanced in the ferromagnetic state. Peak 2 is barely visible at high temperatures but increases its intensity below $T_C$ and saturates before the lowest temperatures are reached. In fact, the temperature dependencies of peaks 2 and 3 are completely symmetric. Another point that has been noticed is that in the more metallic sample at low temperatures ($^{16}$O), peak 2 is accompanied by a narrow Drude peak containing few spectral weight.

In Chapter 6, it was argued that the temperature dependence just described can be encompassed in the phase-separation picture. Below $T_C$ both ferromagnetic and antiferromagnetic phases are present. This is the reason for which peak 3 does not completely disappear in the ferromagnetic state. Moreover, isotope substitution changes the relative volume of one phase in respect to the other. This also explains the fact that in the less metallic sample, peak 3 is more intense at low temperatures.

**Spectral Weight**

A central problem in this thesis was the determination of the region in which the spectral weight contained in the optical conductivity was recovered. It was found that the region where this happens depends on the character of the electrons
involved in low frequency processes. In the silicides and manganites, this happens in a fairly large region as the electrons involved have $d$ character. In the heavy fermion systems studied here, the region was found to be much smaller since the electrons involving the formation of the coherent state are of $f$ character.