Chapter 1

Introduction

The physics of low dimensional quantum antiferromagnets has attracted great interest since the discovery of the high-T_c superconductors by Bednorz and Müller in 1986 [1]. Early on, an intimate relationship between magnetism and the occurrence of high-T_c superconductivity has been suggested by Anderson [2] and the undoped parent compounds of the high-T_c-superconductors are thought to be the best realization of a two-dimensional antiferromagnetic spin-1/2 Heisenberg model. Due to the absence of experimental data, the two-dimensional spin-1/2 Heisenberg antiferromagnet had marked an important unexplored region in quantum statistical mechanics before 1986. Contrary to one dimension, where the effect of quantum fluctuations is so strong to suppress long range order, it has been established by now that the two-dimensional antiferromagnetic spin-1/2 Heisenberg model is characterized by a long range ordered ground state and its low energy excitations are well described in terms of spin waves [3]. For finite temperatures, however, no long range order can exist [4, 5]. Moreover, it is not settled, whether the simple spin-wave picture holds also for high-energy excitations.

As the physics of the two-dimensional cuprates is still not completely understood, it has been suggested to approach it from one dimension, where powerful analytical and numerical techniques are available, and to study the crossover via the ladder systems [6, 7, 8], i.e., by successively increasing the number of coupled one-dimensional chains. Here, the simplest system is the two-leg ladder, which consists of only two coupled chains but already displays striking similarities to the two-dimensional cuprates. In the absence of hole carriers it has a spin gap in the excitation spectrum [9], which resembles the spin-"pseudogap" observed in the underdoped two-dimensional cuprates. In particular, however, theoretical studies have shown that upon doping with holes the ground state of the two-leg ladder becomes dominated by superconducting correlations [9]. For the two-dimensional cuprates, on the other hand, the interplay of magnetism and superconductivity is still unresolved. Moreover, the superconductivity of the ladders is predicted to be in the d-wave channel [10], which is widely accepted to be the relevant symmetry for the two-dimensional cuprates.

Besides the theoretical fascination in the spin-ladders, it has been possible to synthesize ladder compounds like $SrCu_2O_3$ [11] or $(Sr,Ca,La)_{14}Cu_{24}O_{41}$ [12]. The existence of a finite spin-gap in the even-leg ladders has been verified experimentally [13], and in particular superconductivity under high pressure was found in $Sr_{0.4}Ca_{13.6}Cu_{24}O_{41}$ by Uehara *et al.* in 1996 [14]. It has to be added, however, that the origin of superconductivity in the ladder compounds is still actively under investigation and it is not yet clear whether it corresponds to the theoretically predicted superconductivity which is based on the analysis of isolated ladders.

The ladder systems are of interest not only with respect to the occurrence of superconductivity but also with respect to their magnetic properties. Here, the crossover from one to two dimensions is not smooth, because even-leg ladders have a finite spin gap, whereas odd-leg ladders are characterized by a gapless excitation spectrum. In spite of this obvious discrepancy, the even-leg ladders are considered more suitable for studying the crossover to two dimensions [15, 16, 17] (for further discussion see Section 2.3) and are expected to resemble more the two-dimensional system the higher the energy [18].

In this thesis the high-energy spin excitations of the two-dimensional undoped cuprates are of particular interest. This is motivated by the observation of high-energy resonances beyond the two-magnon cut off in the optical conductivity of the two-dimensional undoped cuprates, which cannot be explained within a straightforward spin-wave treatment of the two-dimensional spin-1/2 Heisenberg model. As the two-dimensional problem is difficult to approach, it will be addressed by studying the crossover via the even-leg spin ladders.

Structure of this thesis

Chapter 2 provides an introduction to different approaches to the two-dimensional antiferromagnetic spin-1/2 Heisenberg model and summarizes relevant knowledge about quantum antiferromagnets of even lower dimension, like spin-chains and spin-ladders, which are important with respect to the crossover from one to two dimensions.

The purpose of Chapter 3 is to discuss whether a spin-wave treatment of the two-dimensional spin-1/2 Heisenberg model is able to account for all magnetic properties observed experimentally in the two-dimensional undoped cuprates. First, the derivation of a model Hamiltonian for the two-dimensional cuprates will be recapitulated with a focus on the occurrence of higher order spin interactions like the cyclic four-spin exchange term. Then, the results of several experimental probes for the magnetic properties of the two-dimensional undoped cuprates, like neutron scattering, Raman scattering and the optical conductivity, will be reanalyzed with respect to their compatibility with predictions obtained by a spin-wave treatment of the two-dimensional spin-1/2 Heisenberg model. Theoretical treatments, which include spin-phonon interaction or a cyclic four-spin exchange term will be discussed.

In Chapter 4 the mechanism of phonon-assisted magnetic absorption, which has been suggested by Lorenzana and Sawatzky [19] and which allows the observation of magnetic excitations in the mid-infrared range of the optical conductivity, will be explained. We will generalize it to bilayer compounds, which consist of two coupled CuO_2-planes, and to two-leg spin ladders. Although slightly technical, the purpose of Chapter 4 is to derive explicit formulas for the optical conductivity, which will be used throughout this thesis. Relevant for the optical conductivity are form factors, which will be calculated in perturbation theory in Appendix B, and knowledge of the phonon dispersions, which will be calculated in Appendix D using a shell-model. In Appendix A, the linear response formalism will be recapitulated and the optical conductivity will be expressed in terms of a dipole-dipole moment correlation function.

The magnetic excitations of the two-dimensional undoped cuprates are the subject of Chapter 5. Using interacting spin-wave theory we calculate the phonon-assisted magnetic absorption for a bilayer and compare with the experimental spectra of $YBa_2Cu_3O_6$. For intermediate inter-layer couplings, larger than realized in the cuprate materials, we find a new resonance formed by a quasi bound state of two optical magnons. For realistic inter-layer couplings, we find that the spectrum is dominated by a strong bimagnon resonance but contains only little weight at higher energies, similar to what is observed in the single layer compounds. We include spin-phonon interaction and propose an antibound bimagnon-phonon resonance as a possible origin for the high-energy resonance observed experimentally at the two-magnon cut off. However, a large magnon-phonon interaction is necessary for this process and a comparison with the optical conductivity of a two-dimensional spin-1 antiferromagnet and a one-dimensional spin-1/2 system suggests a purely magnetic origin.

To explore the possible role of strong quantum fluctuations for the high-energy resonance in the two-dimensional undoped cuprates we study similar excitations of two-leg spin ladders in Chapter 6. This approach is supported by the observation of a similar high-energy resonance in the two-leg spin ladder compound $(La,Ca)_{14}Cu_{24}O_{41}$. We use two methods to treat the magnetic excitations of two-leg spin ladders, one an analytical approach, which is based on the Jordan-Wigner transformation, and one numerical technique, the Density Matrix Renormalization Group (DMRG), where we use a program, which was kindly provided by Philipp Brune. The analytical approach is especially suitable to study the formation of a two-triplet bound state, which has recently been observed in the optical conductivity of $(La,Ca)_{14}Cu_{24}O_{41}$ by Marco Windt and Markus Grüninger. The DMRG-correction vector method on the other hand is particularly suitable to investigate the high-energy excitations. For different possibilities of calculating dynamic correlation functions using the DMRG see Appendix F. We identify the dominant contribution to the high-energy continuum excitations and establish its relation to the spin-1 chain.

Due to the similar CuO-plaquette structure of the cuprate spin ladders a cyclic spin exchange of similar magnitude is expected as for the two-dimensional cuprates. We investigate the influence of a cyclic spin exchange term on the magnetic excitations of two-leg spin ladders in Chapter 6 and find that the inclusion of a finite cyclic spin exchange term is indeed necessary to obtain a consistent description of the experimental data. A rich phase diagram has been suggested for a two-leg ladder with cyclic spin exchange term. Here, we resolve some discrepancies present in the literature with respect to the ground state properties for large cyclic spin exchange and discuss the formation of incommensurate spin correlations.

In Chapter 7 the magnetic excitations of four-leg spin ladders are investigated using the DMRG-correction vector method. We discuss the transfer of spectral weight from the former two-triplet bound state of the two-leg ladder to a new resonance at higher energies, which we interpret as a precursor of the bimagnon resonance of the two-dimensional antiferromagnet. We analyze the evolution of the high-energy continuum excitations and discuss the role of the four-leg ladder in the crossover from the two-leg ladder to the two-dimensional antiferromagnet.

Chapter 2

Low Dimensional Quantum Spin Systems

2.1 Two-Dimensional Spin-$\frac{1}{2}$ Antiferromagnet

Before the discovery of the cuprates in 1986, the absence of experimental data for the two-dimensional $S = \frac{1}{2}$-Heisenberg antiferromagnet

$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \mathbf{S}_j \,, \tag{2.1}$$

had marked an important unexplored region in quantum statistical mechanics. Here, $J > 0$ is the exchange coupling and the summation $\langle i, j \rangle$ is restricted to nearest neighbor spins. In three spatial dimensions the ground state of the nearest neighbor Heisenberg model in bipartite lattices is characterized by antiferromagnetic long-range order for all values of the spin quantum number S [20, 21]. For two spatial dimensions, on the other hand, it has been demonstrated by Hohenberg [4] and Mermin and Wagner [5] that no long range order can exist for any finite temperature $T > 0$ when the order parameter is continuous. The situation may be different for the ground state ($T = 0$) and it has been shown that antiferromagnetic long-range order exists for any $S \geq 1$ on a square lattice [22] and on a hexagonal lattice [23]. For $S = \frac{1}{2}$ no rigorous proof is available; there is, however, accumulating evidence for a long-range ordered antiferromagnetic ground state. For a detailed review of different approaches to treat the antiferromagnetic two-dimensional $S = \frac{1}{2}$-Heisenberg model see Manousakis [3].

2.1.1 Quantum Nonlinear σ-Model

An important contribution to the discussion about the ground state of the $S = \frac{1}{2}$-Heisenberg model was a renormalization group analysis of the quantum nonlinear