Summary

In solid state physics, theories based on the concept of the propagation of one single electron in a periodic field due to the nuclei of the atoms and an average Coulomb field due to the other electrons, have been very successful in calculating and predicting a wide range of properties of those solids. Already in an early stage, however, it was noted that some systems have properties that cannot be reconciled with the outcome of one-electron theory calculations. A canonical example of a class of such materials are the transition-metal oxides. Many of these oxides, for instance, are predicted to be metals in single electron theory, but are in fact insulators with large bandgaps. This is due to the fact that electron-electron interactions in these materials cannot be treated on the mean-field level, and in fact the full many problem should be addressed.

The lack of success in treating the full effect of correlations has lead to the development of model Hamiltonians. These model Hamiltonians describe highly idealized systems, where most interactions between the electrons are left out. With a model Hamiltonian one wants to describe only a part of the physical phenomena, which are mostly the low energy phenomena. The total, full energy scale, problem is mapped on a model Hamiltonian that incorporates only some effects on part of the full energy scale.

In the last decades a wealth of materials was discovered where electron correlation seems to play an essential role. Some of these materials exhibit extraordinary properties, phases and phase transitions. Examples are the colossal magneto-resistance (CMR) compounds with magnetic and non-magnetic phases, the high $T_c$ materials with insulating, magnetic, metallic and superconducting phases and the (intercalated) C$_{60}$ compounds with an equally overwhelming richness in phases. Also the longer established Kondo- and heavy fermion systems, low dimensional organic salts and transition metal oxides in general, are dominated by correlation effects. In spite of the enormous efforts, the behavior of these materials is theoretically not well understood.

It would be surprising if all these classes of different correlated systems could be described by one, simple, model Hamiltonian. The possibility exists, however, that the low energy excitations of these systems can be explained in terms of such a simple, effective model. This generates a force-field where the extrema are given by the full many-body problem on one hand and the simplest model Hamiltonian on the other.
In this thesis interactions between electrons that go beyond the simplest model Hamiltonians are included. For the CMR materials the importance of orbital degeneracy is heavily debated and for $C_{60}$ and high $T_c$ compounds the possible relevance of longer range Coulomb interactions is considered. In transition-metal oxides in general and in $C_{60}$ the screening of the Hubbard $U$ is surely important. Model Hamiltonians that incorporate these interactions found their way into this thesis. The primary focus is on the relevance and consequences of the inclusion of these interactions.

**Screening of the Coulomb repulsion**

The question how the electron-electron Coulomb interaction is screened in non-polar insulators is addressed. The screening of the Coulomb repulsion in such a material can be calculated using a point-dipole model. Within the partial continuum limit of this model, where dipole-dipole interactions are taken into account on the Clausius-Mossotti level, the effective on-site Coulomb interaction $U$ can be calculated.

Applying the formalism to $C_{60}$, effective values for the bulk Coulomb interaction can be obtained that agree well with experiment. The effective $U$ for $C_{60}$ on the surface, on metallic substrates and in compounds with ionic $C_{60}$ show that metallic substrates screen $U$ and the nearest neighbor Coulomb interaction $V$ very efficiently. Small lattice contractions can cause a substantial reduction of $U$.

For three dimensional systems the deviations from the Clausius-Mossotti result for the off-site screened Coulomb interaction are small. In one and two dimensional systems, however, only local field effects contribute to the screening. At large distances the Coulomb interaction is unscreened and in 1D at intermediate distances the Coulomb interaction is even anti-screened.

**Inter-site Coulomb repulsion**

In an extended Hubbard model with an inter-site Coulomb repulsion $V$ at half filling, $V$ does not screen the correlation gap but introduces charge neutral excitations inside the gap.

For single band Hubbard models, the results suggest an enhancement of the exchange constant, roughly as $J(V) = 4t^2/(U - V)$, whereas the spin dynamics still remains consistent with that of an ideal Heisenberg antiferromagnet. The remarkable feature of the results is that this rescaling seems to hold up to the CDW instability, which opens the way to unexpectedly large values of the ratio $J/t$. In particular, for the strong correlation case close to a CDW instability in 1D, the results suggest values of $J$ comparable to the original hopping integral. Therefore, real materials which may be described by a strong correlation single band Hubbard model close to a CDW instability may provide realizations of $t-J$-like models with ‘un-physically’ large values of $J/t \approx 1$.

For charge transfer systems, the super-exchange via ligand orbitals on the contrary is suppressed by the inter-site Coulomb repulsion because the nearest neighbor repulsion $V$ increases the ‘bare’ charge transfer energy. If the charge transfer gap
is kept constant while introducing $V$, the super-exchange increases because $V$ introduces low energy excitonic states. This might be relevant for theoretical models of cuprate superconductors which rely on large values of $V$.

The features of a Hubbard model with inter-site Coulomb interactions upon doping or at finite temperature are both qualitatively and quantitatively different from the simple Hubbard model. In the excitation spectrum at finite doping, the nearest neighbor Coulomb interaction introduces satellites both in the high and low energy part of the spectrum and renormalizes the pseudo gap. It increases the low energy spectral weight. At finite temperatures states inside the gap obtain spectral weight, and the gap seems to fill in gradually. Excitonic states in the insulating undoped system visible only in optical spectroscopies attain an effective charge in the doped systems and become visible in the electron removal and addition spectra. These states quickly fill in the gap with spectral weight especially in higher dimensions. The inclusion of the nearest neighbor Coulomb interactions in the case of $C_{60}$ may provide an explanation for the large spread in the photoemission spectra.

**Effective Hubbard Hamiltonian**

A description by means of Hubbard-like Hamiltonians, using renormalized parameters, may yield misleading results. When screening effects are taken into account explicitly, the conductivity gap in the spin density wave regime is determined by the screened on-site Coulomb repulsion and independent of nearest neighbor Coulomb repulsion. The point, however, at which the transition from the spin density wave to the charge density wave regime takes place, is determined by the bare values of on-site and nearest neighbor Coulomb interactions. In the phase diagram for the interaction part of the extended Hubbard model including polarization screening two new phases, one of which is ferroelectric, and a $U = 0$ phase transition appear. The $U = 0$ phase transition between two different types of charge density waves shows that the effect of a nearest neighbor Coulomb interaction and a polarizability are quite different. Furthermore polarization screening tends to drive the SDW-CDW phase transition from 1st to 2nd order. This may serve as an example of the fact that in predicting phase transitions in the various Hubbard models, screening cannot be taken into account by using effective parameter sets, but that screening mechanisms should be explicitly incorporated in the model Hamiltonian.

**Degeneracy**

A model Hamiltonian that captures the low energy behavior of a two-fold degenerate Hubbard Hamiltonian with Hund’s rule coupling, is studied. The phase diagram in the mean-field limit and in a two-particle approach reveals a rich variety of phases where both the orbital degrees of freedom and the spin degrees of freedom can be ordered (anti-)ferromagnetically. In this case there may exist, besides usual spin-waves (magnons) also orbital-waves (orbitons) and, most interestingly, the combined spin-orbital excitation which can be visualized as bound states of magnons and orbitons. Both in one- and two-dimensions the bound states are the lowest lying elementary
excitations, much lower than the single spin or single orbital excitations. This shows that the elementary excitations in orbitally degenerate strongly correlated electron systems in general carry both spin and orbital character.

Optical conductivity of $A_3C_{60}$

The optical conductivity of $A_3C_{60}$ is calculated, including the effects of the lowest order self-energy diagram due to the electron-phonon interaction. This coupling reduces the width of the Drude peak and transfers weight to the mid-infrared peak at an energy somewhat larger than the phonon frequency. This leads to a mid-infrared peak with an energy of the right order of magnitude, but probably a bit too large. The reduction of the width of the Drude peak goes in the right direction, but it is much too small. These are all strong indications that for understanding the properties of $A_3C_{60}$ electron-phonon interactions are not sufficient.

t-t'-'-J model

For the two dimensional $t - t' - t'' - J$ model at finite temperature a technique to deal with the state without long range antiferromagnetic order was developed. There is hope to extend this technique to the spin liquid state of the doped system. The self-consistent Born approximation is generalized to the case of nonzero temperature and the Dyson equation which relates the single hole Green’s function with fixed spin to the single hole Green’s function with fixed pseudospin is derived. This equation is sensitive to very large distances of order of the magnetic correlation length and therefore not convenient for computations. To overcome this problem a renormalization procedure is developed which allows one to exclude large distances and to work with a relatively small lattice.

The calculated ARPES spectra demonstrate that temperature broadening is not enough to explain widths of the experimental spectra. This strengthens the argument that other degrees of freedom contribute to the peak width.