Chapter One

Introduction

1.1 Nonlinear Excitations

Excitations in one-dimensional (1-D) magnets with Heisenberg exchange traditionally have been described by small perturbations from the ground state, that is, linearized modes called spin waves classically or magnons when quantized. This spin wave interpretation more recently has been challenged by a broader interpretation which includes nonlinear excitations. The experiments supporting this new interpretation include neutron scattering, specific heat, and NMR relaxation time measurements, on materials such as $CsNiF_3$ and $(CH_3)_4NMnCl_3$ (TMMC). From the classical viewpoint, it is apparent that spin waves cannot serve as a complete basis set of excitations for the low temperature thermodynamics, since 1-D magnetic Heisenberg Hamiltonians generally have a much richer spectrum of nonlinear large amplitude excitations in addition to the spin waves. These large amplitude classical excitations, called solitons or kinks, can topologically connect degenerate ground states, making it impossible for them to be expressed as a superposition of linear spin waves. Similarly one might expect there to be analogous nonlinear quantum mechanical excitations that cannot be expressed as multi-magnon states. Only a solution to the full nonlinear equations of motion can give these nonlinear modes. Here we investigate the classical dynamics of these excitations, both by numerical methods and analytic calculations, and the quantum statistical mechanics of these systems by a quantum Monte Carlo method. Also we will investigate the possibility of 1-D magnets as being model systems for solid state chaos, as exhibited by complexity in the time and space response of the damped driven system.

1.2 Classical Kink Dynamics

The model spin Hamiltonian studied is the easy-plane anisotropic Heisenberg Hamiltonian, with an applied symmetry-breaking field in the easy plane. Ferromagnetic and antiferromagnet exchange J will be considered. It has been shown (Mikeska and Patzak 1977, Bishop 1980) that classical mechanics should apply for $S \gg (A/2\pi^2 J)^{1/2}$, where S is the spin and A is the easy-plane anisotropy constant. (See the Hamiltonian in equation (2-1)). This condition is met fairly well for some real materials, notably the ferromagnets $CsNiF_3$ (S = 1) and CHAB ($(C_6H_{11}NH_3)CuBr_3$, $S = \frac{1}{2}$), and the antiferromagnet TMMC ($(CH_3)_4NMnCl_3$, $S = \frac{5}{2}$). Therefore an investigation of the classical dynamics for this Hamiltonian, especially for the nonlinear modes, should have relevance for experiments. Furthermore, when exact simple solutions are not known, it is easier to study the classical dynamics using numerical simulations or Ansatz calculations and then quantize semiclassically rather than perform exact diagonalization of the quantum many particle Hamiltonian. Classical dynamics is the subject of Part I of this thesis.

The dynamics of the classical spin vectors has been mapped approximately to the sine-Gordon (sG) equation for both the ferromagnet and the antiferromagnet (Mikeska 1978, 1980, Flüggen and Mikeska 1983). Requirements for this mapping to be applicable are small applied field and strong easy-plane anisotropy compared to the exchange, which combine to restrict the spins to the easy plane, allowing only small out-of-plane motions. For the ferromagnet, the kink is a rotation of the of the spin vectors through 2π as one moves down the chain. Taking the kinks to be right-handed screwlike rotations of the spin vectors, one can also have left-handed rotations, called antikinks. For the antiferromagnet, the rotation is through π, connecting the two physically distinct degenerate ground states. In both cases the kink is a traveling wave, with a fundamental creation or rest energy. This creation energy is the energy of the static (zero velocity) kink. Sine-Gordon theory predicts that the kink energy increases monotonically with velocity, in the sense of a relativistic increase in mass (see for example Scott et al. 1973, Bishop et al. 1980).

The static ferromagnetic kinks were shown to have an energetic instability when the field is raised above a critical field -- a localized mode could be excited by which the kink could lower its energy (Kumar 1982, Magyari and Thomas 1982). The mode involves a tipping of the spins up out of the easy plane, thereby invalidating the assumptions which led to the sG equation. Then a more complete analysis is necessary, using the full Hamiltonian without any sG approximations. One simple way to obtain dynamic information, especially to answer questions about kink stability outside the sG regime, is by numerical integration of the equations of motion on a discrete lattice. Presently there is no other way to test the stability of moving kinks. These numerical simulations, to be described, also give the single kink energy as a function of velocity and applied field, showing strong deviations from ideal sG behavior. In particular, at fields greater than the critical field, dynamically stable kinks with negative effective masses are possible, which should go over into the known pulse solitons of the isotropic Heisenberg model as the field is increased to values large compared to the anisotropy field. These simulations also have been used to study kink-antikink scattering, further emphasing strongly non-sG behavior.

A distinction is made here between energetic and dynamic linear stability. Energetic linear stability analysis involves linearizing the Hamiltonian about the sG kink solution, and looking for modes by which the kink can lower its energy, by solving a time-independent eigenvalue problem. In a dynamic linear stability analysis, the full nonlinear equations of motion are linearized about the sG kink solution, leading to time-dependent dynamic equations of motion. Existence of modes growing with time then indicates an instability which is dynamically allowed. The energetic stability analysis will be seen to predict a ferromagnetic kink instability that is not found in the dynamic stability equations, nor in the numerical simulations. To be more precise, there is no structural instability in these excitations above the critical field, but there is behavior that cannot be described by moderately perturbed sG theory. Even for fields less than the critical field, the instability is manifested in the energy dispersion of the kinks, causing the energy to become a double valued function of velocity.

For the ferromagnet kink-antikink ($K\bar{K}$) collision simulations an interesting variety of possible output states exists, depending on the applied field and the incoming velocity of the pair. Although it might be expected that the $K\bar{K}$ pair would transmit through each other in a sG manner for fields below the critical field, this is not necessarily the case. At very low fields and moderate velocities sG-like transmission does occur. For higher fields below the critical field, the pair form an oscillatory breather-like mode, and for higher fields, but still below the critical field the pair reflect. For applied fields greater than the critical field, where the single kinks always have a negative effective mass, the pair has always been seen to reflect. Of course at still larger fields, the kink width would become smaller and discreteness effect may modify this result. Generally, then, it has been found that sG behavior in $K\bar{K}$ collisions tends to be the exception rather than the rule. This is not necessarily inconsistent with the results of experiments on real materials, especially inelastic neutron scattering for $CsNiF_3$, where kinks and breathers (bound $K\bar{K}$ pairs with an internal oscillation frequency) may have similar signatures.

For the antiferromagnetic kinks, Flüggen and Mikeska (1983) have shown that there are generally two branches possible, called xy and yz kinks, depending on the plane in which the spins rotate. At fields small compared to the anisotropy, it is energetically favorable for the spins to rotate around the anisotropy axis, thereby minimizing the anisotropy energy. At fields large compared to the anisotropy, it is energetically favorable for the spins to rotate around the applied field direction, thereby minimizing the magnetic field energy. There is a crossover critical field where the two static kink energies are equal, and one might expect that generally the lower energy kink should be the only stable one. Again numerical simulation is the quickest way to test the stability. Doing so, it has been found that the xy branch has an instability similar to that seen in the ferromagnetic kinks (i.e. strongly non-sG behavior), and that the yz branch kinks require a minimum applied field for stability. This yz stability field depends on velocity. These results for yz kinks are also predicted from a linear dynamic stability analysis given here. The numerical results are in good agreement with the analytic predictions.

Further information about the exact dynamical kink solutions for these 1-D magnets can be obtained by making various kink Ansätze. Some previous Ansätze for the ferromagnetic kinks (Kumar 1982, Magyari and Thomas 1982, Liebmann et al. 1983) are compared with the numerical simulation results, and it is seen that the one by Liebmann et al. presently is in closest agreement with the simulations. This Ansatz works especially well compared to the others because it is based on geometrical intuition, and because it is formulated in xyz spin components rather than spherical coordinates, which can have problems near the polar axis. The success of this type of Ansatz for the ferromagnet suggested modifying the calculation to make it applicable to the antiferromagnetic kinks. This is presented in Chapter Six. The results obtained are in good agreement with numerical simulations. But an important feature of the Ansatz is that it characterizes the xy and yz kinks as being closely related geometrically, and that they both can be considered as belonging to a single energy dispersion curve.

Finally one further aspect of the classical spin dynamics considered here is the effect of driving the system with combined DC plus AC applied magnetic fields, together with the addition of a phenomenological damping term to the equations of motion. In particular for the ferromagnet, the case of a DC field in the easy plane plus an AC field perpendicular to the easy plane is shown to be equivalent to a damped driven sG equation in the small out-of-plane angle approximation. Considerable numerical work (Bishop 1984) has shown that this equation and ones similar to it have a rich space time structure, including, coexisting spatial coherence and temporal chaos in appropriate parameter regimes. Similar results are found here for the full nonlinear ferromagnet equations, in the damped driven sG limit and for other field configurations.

It is concluded from these studies that simple sG theory for classical 1-D planar magnets is typically inadequate, especially due to the existence of kink instabilities. These instabilities are driven by the extra out-of-plane degree of freedom which is too strongly suppressed in the sG limit. This has implications for the thermodynamics as given by classical phenomenological ideal gas theory, where weakly interacting longlived, robust soliton excitations are assumed. Certainly the results from the $K\bar{K}$ scattering simulation indicate that sG phenomenological theory must be modified to be applicable here.

1.3 Spin Statistical Mechanics

Classical statistical mechanics for these easy-plane ferromagnets has been investigated by many others from the point of view of transfer matrix approaches and the approximately equivalent sG soliton-phonon gas phenomenological theory. It is now clear that the pure sG ideal gas theory cannot be fitted to available experimental data (for specific heat of CHAB and $CsNiF_3$) without at least a renormalization of the soliton rest energy. Even with the renormalization the agreement between theory and experiment is good only for some quantities, such as the positions of the peaks in the specific heat verses field as the temperature is varied. The heights of these peaks fit poorly (Ramirez 1984). This renormalization presently seems to have two possible explanations; one from including out-of-plane motions in the classical transfer matrix that were excluded from the sG limit (Mikeska 1978, Kumar and Samalam 1982), the other from performing a quantization of the classical sG equation (Maki and Takayama 1979, Maki 1980, 1981, Mikeska 1982). For $CsNiF_3$, the classical transfer matrix approach leads to approximately the same kink energy reduction ($\gtrsim$ 20%) as does the quantized sG theory. A similar reduction is necessary to fit classical sG theory to experiment. However, it seems that both of these methods are flawed. Recent evidence from a numerical transfer matrix calculation for the full Hamiltonian (Pini and Rettori 1984) shows that a classical description for $CsNiF_3$ is not adequate. Quantum effects must therefore be important. But taking the classical sG equation limit of the full equations of motion, and then quantizing the sG equations seems to be logically flawed, especially when one knows that the out-of-plane classical spin motions make the mapping to sG theory invalid except for a small range of parameters. Quantum mechanics should be included from the start.

The problem of classical verses quantum statistical mechanics for the easy-plane ferromagnet is studied here by using a quantum Monte Carlo method suggested by Suzuki (1976), and tested for $S = \frac{1}{2}$ by Cullen and Landau (1983) and Wiesler (1982) and others. The method converts the problem of finding the partition function for a 1-D quantum system to that of finding the partition function for a 2-D classical system. The 2-D classical system is an n vertex model. For instance, the spin-$\frac{1}{2}$ fully anisotropic quantum Heisenberg model is equivalent to an 8 vertex model (Baxter 1972). The mapping is approximate since it uses a path integral in discrete inverse temperature steps to evaluate the quantum partition function, rather than a continuous path integral where the inverse temperature steps go to zero. The method for $S = \frac{1}{2}$ has been modified to make it applicable to S = 1 systems. The complexity of implementation by computer scales with the spin value S; only $S = \frac{1}{2}$ and S = 1 calculations are presented here. Indeed for higher values of S a classical description is probably adequate -- for instance, the $S = \frac{5}{2}$ antiferromagnet TMMC is a good example. These calculations are presented for comparison with the phenomenological sG ideal gas theory and with available experimental data for CHAB and $CsNiF_3$.

Definitive conclusions about the existence of quantum kink excitations in low spin systems are difficult to make based on the thermodynamics as derived from quantum Monte Carlo simulations. However, these studies should throw further light on whether classical or quantum statistical mechanics is necessary to describe the experimentally observed behavior in low spin 1-D planar magnets.

1.4 Summary of Experimental Status: $CsNiF_3$, CHAB and TMMC

The results of a number of experiments on $CsNiF_3$ have been interpreted in terms of a classical gas of sG solitons plus spin waves. These include inelastic neutron scattering (Kjems and Steiner 1978, Steiner et al. 1976, 1983), NMR relaxation time (Goto and Yamaguchi 1981), optical absorption (Cibert and d'Aubigné 1981) and specific heat measurements (Ramirez and Wolf 1982). Since these experiments, however, serious objections have been raised against the soliton interpretation, most importantly that the neutron scattering results could more likely be explained in terms of multimagnon difference processes (Loveluck, Schneider, Stoll and Jauslin 1980-82, Reiter 1981). Also, the fit of the specific heat data is qualitative only; it is obvious that something other than pure classical sG theory is necessary to model $CsNiF_3$. Results here, especially the $K\bar{K}$ scattering simulations, furthermore show that interpretations in terms of modified sG solitons may also be incorrect. Assuming that the model Hamiltonian and parameters as described in Chapter Two are indeed applicable, the question is: what assumptions about the dynamics are necessary to improve the agreement between theory and experiment? Currently, there are major controversies concerning the necessity of quantum versus classical mechanics, out-of-plane spin motions versus pure sG theory and kinks versus multimagnon states. The purpose here is to provide further theoretical information to help to decide these questions.

For the spin-½ magnet CHAB, the experimental knowledge is less developed. Although the physical properties are well characterized (see section 2.1), currently the only experimental evidence for sG solitons comes from specific heat and NMR measurements (Kopinga et al. 1984). Here, as for $CsNiF_3$, the fit for the specific heat is qualitative only. However, there have been ferromagnetic resonance experiments on the clorine isomorph CHAC (Hoogerbeets et al. 1984) which have been interpreted in terms of magnon bound state excitations, and this could be an alternative interpretation for the CHAB experiments.

Probably TMMC is the best example where the experimental evidence for solitons is convincing. The physical properties are well characterized, as described in section 5.1. A number of experiments have provided evidence for the in-plane antiferromagnetic kinks, including inelastic neutron scattering (Boucher et al. 1980, 1981, Regnault et al. 1982), NMR relaxation time measurements (Boucher and Renard 1980), field dependence of the Néel temperature (Boucher 1980), and specific heat (Borsa et al. 1983), provided the kink creation energy is assumed to be renormalized due to quantum effects. The agreement between experiment and sG theory is generally good in these experiments, and can be improved somewhat by including effects of the out-of-plane kinks (Harada et al. 1981). The major purpose here, then, is to further investigate the properties of the out-of-plane kinks, especially their stability as a function of the applied field.