THERMODYNAMICS of QUANTUM SPIN CHAINS

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1. Introduction and summary.

In this paper we consider the ferromagnetic chain with easy-plane anisotropy D and in-plane applied Zeeman field H. Its quantum hamiltonian reads:

$$\hat{\mathcal{H}} = \sum_{i=1}^{N} \left[-J\hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+1} + D\left(\hat{S}_i^z\right)^2 - g\mu_B H \hat{S}_i^x \right] \quad , \tag{1.1}$$

with $\left|\hat{\mathbf{S}}\right|^2 = S(S+1)$. This hamiltonian is believed to have a physical realization in the real system $CsNiF_3$, with $S = 1$, $J = 23.6$ K, $D = 9$ K, $g = 2.4$.

For strong planar anisotropy, the classical counterpart of (1.1) can be approximated by the so-called planar model, which in the continuum limit becomes the sine-Gordon (SG) model. The latter admits soliton excitations contributing to the thermodynamics. This has lead to the search for soliton-like behavior in the easy-plane ferromagnetic chain, both theoretically [1,2] and experimentally [3–5]. Indeed, experimental measurements [4] of the magnetic contribution to the specific heat in $CsNiF_3$ have risen many attempts to relate the peak in the *excess* specific heat (i.e. the difference between the specific heat with and without applied field H) with the existence of SG-like nonlinear excitations [5,6].

However, as it has been clearly shown by Pini and Rettori [7], the thermodynamics of $CsNiF_3$ requires a quantum mechanical treatment, especially in order to account for the relevant quantum character of the out-of-plane fluctuations [5–10]. Quantum corrections to the classical SG thermodynamics have been introduced [5,8,9], and modified SG-like theories have been developed [6,10] in order to include a better description of the out-of-plane part.

In this paper we put forward a new approach. We start by observing that the quantum hamiltonian (1.1) can be expressed in terms of canonical operators by means of the Villain transformation [11]. Then, in section 2 we show how to approximate the general path-integral formula for the partition function [12,13] by using a nonlocal quadratic "trial" action functional, containing a number of variational parameters which are functions defined in phase-space. These parameters can be at best determined after a variational principle, and the approximated path-integral can be put into the classical form of a phase-space integral, allowing us to define an effective hamiltonian. In order to

get simple and useful expressions, we then introduce an approximation of "low quantum coupling", which, however, preserves an exact description of the quantum behavior of the harmonic part — even at lowest temperatures, where the results of the selfconsistent gaussian approximation are actually recovered —, as well as of the classical nonlinear behavior — with quantum renormalizations which, in the high temperature limit, do agree with the results of the Wigner method —.

In section 3 we apply the above framework to the (Villain transformed) hamiltonian (1.1). We show that the corresponding effective hamiltonian can be recast, in terms of classical spin variables, into the same form of the original hamiltonian, with suitably renormalized parameters, but for the appearance of an exchange anisotropy term. Then we use the effective hamiltonian in numerical transfer matrix calculations, and report some quantum results for the specific heat, showing their fairly good quantitative agreement with the available experimental data for $CsNiF_3$, as well as with computational data by other authors [14]. In addition, we also compare with the results of the quantum planar and SG models, for which a simplified version of the variational method leads to the corresponding effective potentials [9,10], showing that the nonlinearity of the exchange term (which is neglected in SG) plays a relevant role. We conclude that the nonlinear thermodynamic behavior of the spin chain only qualitatively can be explained in terms of SG solitons.

2. The effective hamiltonian.

Let $\hat{\mathcal{H}}(\hat{p},\hat{q})$, be the hamiltonian operator of a quantum mechanical system with N degrees of freedom. We use the matrix notation $\hat{p} = \{\hat{p}_i\}_{i=1,\dots,N}$ and $\hat{q} = \{\hat{q}_i\}_{i=1,\dots,N}$ for its momentum and coordinate operators, which satisfy canonical commutation relations $[\hat{p}_i, \hat{q}_j] = i\hbar\delta_{ij}$.

The equilibrium partition function $\mathcal{Z}$ of this system at the temperature $T = \beta^{-1}$ can be expressed as a functional integral over paths $\{p(u), q(u)\}$ in phase-space:

$$\mathcal{Z} \equiv e^{-\beta F} = \int \mathcal{D}[p(u)] \int\limits_{q(0)=q(\beta)} \mathcal{D}[q(u)]\ e^{\,S[p(u),q(u)]} \quad , \tag{2.1}$$

with the action functional

$$S[p(u),q(u)] = \int_0^{\beta\hbar} \frac{du}{\hbar} \left[ip^t\dot{q} - \mathcal{H}(p,q)\right] \quad . \tag{2.2}$$

In order to give a mathematical meaning [12] to the above formulas, one has to define the function $\mathcal{H}(p,q)$, which is associated with the hamiltonian operator $\hat{\mathcal{H}}(\hat{p},\hat{q})$ by some ordering rule (e.g. the p-q order [12,13], obtained by moving the $\hat{p}_i$ on the left of the $\hat{q}_i$), and an additional condition on the $p^t\dot{q}$ term (which, in the case of p-q order, reads $p^t\dot{q} \to p^t(u)\dot{q}(u+0)$).

If we are able to calculate path-integrals with a trial action $S_0[p(u),q(u)]$, containing a proper set of variational parameters [13], it is possible to minimize the r.h.s. of the so-called Feynman-Jensen inequality:

$$F \leq F_0 + \beta^{-1}\langle S_0 - S\rangle_{S_0} \quad , \tag{2.3}$$

in order to approximate (2.1). The functional average $\langle\ \cdot\ \rangle_{S_0}$ appearing in (2.3) corresponds to the path integral (2.2) with the integrand functional e^{S_0} multiplied by $S_0 - S$, divided by $\mathcal{Z}_0 = \exp(-\beta F_0)$.

The trial action we will use has the following general form:

$$S_0[p(u),q(u)] = \int_0^{\beta\hbar} \frac{du}{\hbar}\left[ip^t\dot{q} - w - \frac{1}{2}\delta p^t\ \mathbf{A}^2\ \delta p - \delta p^t\ \mathbf{X}\ \delta q - \frac{1}{2}\delta q^t\ \mathbf{B}^2\ \delta q\right] \quad , \tag{2.4}$$

where $\delta p \equiv p(u) - \bar{p}$ and $\delta q \equiv q(u) - \bar{q}$, and the functionals

$$(\bar{p},\bar{q}) = \int_0^{\beta\hbar} \frac{du}{\beta\hbar}\ (p(u),q(u)) \tag{2.5}$$

represent the *average point* of each path $(p(u),q(u))$. Moreover, the real c-number w and the real $N \times N$ matrices $\mathbf{A}$, $\mathbf{X}$ and $\mathbf{B}$ are functions of $(\bar{p},\bar{q})$, and are considered as the variational parameters of the trial action. Of course, they are subjected to proper constraints (for instance, $\mathbf{A}$ and $\mathbf{B}$ are symmetric). In addition, the matrix $\mathbf{\Sigma} \equiv \mathbf{A}^{-1}\mathbf{X}\mathbf{A}$ is constrained to commute with the matrix $\mathbf{B}$, which makes the path integrals with S_0 appearing in (2.3) of easy evaluation. The final result for $\mathcal{Z}_0$ can be put in the classical form of a phase space integral over $\bar{p}$ and $\bar{q}$ (in the forthcoming formulas we will suppress the bars).

The calculation leads to the formal diagonalization problem of the matrices $\mathbf{\Sigma}$ and $\mathbf{\Omega}^2 \equiv \mathbf{A}\mathbf{B}^2\mathbf{A} - \mathbf{\Sigma}^t\mathbf{\Sigma}$, which are diagonalized by the orthogonal matrix $\mathbf{U}(\bar{p},\bar{q})$:

$$\sigma_k\ \delta_{kl} = \left(\mathbf{U}\ \mathbf{\Sigma}\ \mathbf{U}^t\right)_{kl} \quad , \qquad \omega_k^2\ \delta_{kl} = \left(\mathbf{U}\ \mathbf{\Omega}^2\ \mathbf{U}^t\right)_{kl} \quad , \tag{2.6}$$

and we can equivalently consider as variational parameters to be determined by (2.3) the independent components of the matrices $\mathbf{A}(p,q)$ and $\mathbf{U}(p,q)$, the sets of eigenvalues $\{\omega_k^2(p,q)\}$ and $\{\sigma_k(p,q)\}$, and the function $w(p,q)$. The minimization of the r.h.s. of (2.3) with respect to w yields the vanishing of $\langle S - S_0\rangle_{S_0}$, so that we are allowed to define an effective hamiltonian $\mathcal{H}_{\rm eff}(p,q)$ by which the approximated partition function can be written in the classical form

$$\mathcal{Z}_0 \equiv e^{-\beta F_0} = \int \frac{dp\ dq}{2\pi\hbar}\ e^{-\beta\mathcal{H}_{\rm eff}(p,q)} \quad , \tag{2.7}$$

$$\mathcal{H}_{\rm eff}(p,q) = e^{\langle\!\langle\phi^2\rangle\!\rangle}\ \mathcal{H}(p,q) - \sum_k\ \omega_k^2\ \alpha_k + \frac{1}{\beta}\sum_k\ \ln\frac{\sinh f_k}{f_k} \quad , \tag{2.8}$$

where

$$f_k(p,q) = \frac{1}{2}\beta\hbar\omega_k \ , \qquad \alpha_k(p,q) = \frac{\hbar}{2\omega_k}\left(\coth f_k - f_k^{-1}\right) \quad . \tag{2.9}$$

In eq. (2.8) $\mathcal{H}(p,q)$ represents the function associated with $\hat{\mathcal{H}}$ by Weyl ordering [12], also called the "Weyl symbol for $\hat{\mathcal{H}}$":

$$\mathcal{H}(p,q) = \int ds \left\langle q - \frac{s}{2} \right| \hat{\mathcal{H}}(\hat{p},\hat{q}) \left| q + \frac{s}{2} \right\rangle e^{ip^t s/\hbar} \quad , \tag{2.10}$$

and the differentiation operator $\hat{\phi} \equiv (\eta^t \partial_p + \xi^t \partial_q)/\sqrt{2}$ acts on the arguments of $\mathcal{H}(p,q)$. The gaussian average $\langle\!\langle \cdot \rangle\!\rangle$, which operates on the fluctuation variables $\{\eta_i\} = \eta = \mathbf{A}^{-1}\mathbf{U}^t\tilde{\eta}$ and $\{\xi_i\} = \xi = \mathbf{A}\mathbf{U}^t\tilde{\xi}$, is uniquely defined by the second moments of their transformed components $\{\tilde{\eta}_k\}$ and $\{\tilde{\xi}_k\}$:

$$\langle\!\langle \tilde{\eta}_k^2 \rangle\!\rangle = (\omega_k^2 + \sigma_k^2)\alpha_k \quad , \qquad \langle\!\langle \tilde{\eta}_k \tilde{\xi}_k \rangle\!\rangle = -\sigma_k \alpha_k \quad , \qquad \langle\!\langle \tilde{\xi}_k^2 \rangle\!\rangle = \alpha_k \quad . \tag{2.11}$$

The further minimization of F_0 with respect to the remaining parameters gives their determination as

$$\begin{aligned} \mathbf{A}_{ij}^2 &= e^{\langle\!\langle \hat{\phi}^2 \rangle\!\rangle}\, \mathcal{H}_{p_i p_j}(p,q) \quad , \\ (\omega_k^2 + \sigma_k^2)\, \delta_{kl} &= \sum_{ij} (\mathbf{U}\mathbf{A})_{ki} \left(e^{\langle\!\langle \hat{\phi}^2 \rangle\!\rangle}\, \mathcal{H}_{q_i q_j}(p,q) \right) (\mathbf{U}\mathbf{A})_{lj} \quad , \\ \sigma_k\, \delta_{kl} &= \sum_{ij} (\mathbf{U}\mathbf{A}^{-1})_{ki} \left(e^{\langle\!\langle \hat{\phi}^2 \rangle\!\rangle}\, \mathcal{H}_{p_i q_j}(p,q) \right) (\mathbf{U}\mathbf{A})_{lj} \quad . \end{aligned} \tag{2.12}$$

These are coupled self-consistent secular equations, the first of which determines the "reciprocal mass" matrix $\mathbf{A}^2$. The subscripts of $\mathcal{H}$ denote the corresponding derivatives.

The above formalism, when applied to a quadratic hamiltonian (with the "mixed" term satisfying the above mentioned constraint of commutativity), gives the exact partition function. Indeed the variational parameters turn out to be constant, and equal to the corresponding coefficients in the starting hamiltonian, so that the partition function turns out to be the fully quantum one, $\mathcal{Z} = \sum_k \left(2 \sinh f_k\right)^{-1}$, thanks to the logarithmic term appearing in $\mathcal{H}_{\text{eff}}$.

However, it is generally very hard to solve the above self-consistent equations for any phase-space point, a task that presumably will numerically last as a heavy quantum Monte Carlo simulation. Therefore a further simplification is in order. If the operator $e^{\langle\!\langle \hat{\phi}^2 \rangle\!\rangle}$ only slightly affects the Weyl hamiltonian, we can rederive the "low coupling approximation" (LCA) that we have extensively used in the standard case of the effective potential [9,13] .

The LCA consists in expanding the variational parameters around their values in the self-consistent minimum (p_0, q_0) of $\mathcal{H}_{\text{eff}}(p,q)$, in such a way that $\mathcal{H}_{\text{eff}}$ is correct within terms of order α_k^2. The simplest form of the LCA occurs in the case of translationally invariant systems, for which the matrix $\mathbf{U}(p_0,q_0)$ is nothing else but a standard real Fourier transformation, which also diagonalizes the reciprocal mass matrix:

$$\left(\mathbf{U}\mathbf{A}^2\mathbf{U}^t\right)_{kl} = m_k^{-1}\, \delta_{kl} \quad . \tag{2.13}$$

Here and in the following the dependence on (p_0, q_0) is understood. The LCA effective hamiltonian then reads

$$\mathcal{H}_{\rm eff}(p,q) = e^{\langle\!\langle \phi^2 \rangle\!\rangle}\, \mathcal{H}(p,q) - \langle\!\langle \phi^2 \rangle\!\rangle\, e^{\langle\!\langle \phi^2 \rangle\!\rangle}\, \mathcal{H}(p_0,q_0) + \frac{1}{\beta}\sum_k \ln \frac{\sinh f_k}{f_k} \quad , \tag{2.14}$$

and the variational parameters can be obtained by Fourier transformig the second derivatives of $\mathcal{H}_{\rm eff}$

$$\begin{aligned} m_k^{-1} &= \sum_{ij} U_{ki}U_{kj}\partial_{p_i}\partial_{p_j}\mathcal{H}_{\rm eff}(p,q)\Big|_{p_0,q_0} \quad , \\ \sigma_k &= \sum_{ij} U_{ki}U_{kj}\partial_{p_i}\partial_{q_j}\mathcal{H}_{\rm eff}(p,q)\Big|_{p_0,q_0} \quad , \\ m_k\left(\omega_k^2+\sigma_k^2\right) &= \sum_{ij} U_{ki}U_{kj}\partial_{q_i}\partial_{q_j}\mathcal{H}_{\rm eff}(p,q)\Big|_{p_0,q_0} \quad . \end{aligned} \tag{2.15}$$

It turns out that the first term of the effective hamiltonian is the result of a broadening of the Weyl hamiltonian, on a scale given by eqs.(2.11) for each "normal mode". Eqs.(2.11) represent the pure quantum contributions (i.e. the quantum ones minus the corresponding classical) to the square fluctuations of the canonical coordinates for a hamiltonian corresponding to the quadratic approximation to $\mathcal{H}_{\rm eff}$. Finally, we note that the high temperture limit of $\mathcal{H}_{\rm eff}$ concides with the Wigner effective hamiltonian, so that the Weyl ordered hamiltonian $\mathcal{H}$ represents the *well-done* classical limit.

3. Effective hamiltonian of the spin chain.

Exploiting the easy-plane character of the system (1.1), we perform, for each spin operator $\hat{\mathbf{S}}$, the quantum Villain transformation [11] to canonical coordinate operators $\hat{\mathbf{S}} \longrightarrow \{\hat{S}^z, \hat{\varphi}\}$, satisfying $[\hat{\varphi}, \hat{S}^z] = i$:

$$\hat{S}^+(\hat{S}^z,\hat{\varphi}) = e^{i\hat{\varphi}}\,\sqrt{S(S+1)-\hat{S}^z(\hat{S}^z+1)} \quad , \qquad \hat{S}^- = (\hat{S}^+)^\dagger \quad . \tag{3.1}$$

The Villain transformation allows us to use the variational method described in the preceding section. The Weyl symbol for $\hat{S}^\pm(\hat{S}^z,\hat{\varphi})$ is easily found to be:

$$S^\pm(S^z,\varphi) = \sqrt{\left(S+\frac{1}{2}\right)^2-\left(S^z\right)^2}\; e^{\pm i\varphi} \quad , \tag{3.2}$$

It follows that it is natural to scale the "momenta" $\hat{S}_i^z$ with $\widetilde{S} = S + \frac{1}{2}$, defining $\hat{p} = \hat{S}^z/\widetilde{S}$. Then $[\hat{\varphi},\hat{p}] = i/\widetilde{S}$, and $\widetilde{S}^{-1}$ plays the role of $\hbar$. Eventually, the Weyl

symbol for $\hat{\mathcal{H}}(\hat{p},\hat{\varphi})$ reads

$$\mathcal{H}(p,\varphi)= J\widetilde{S}^2\sum_i\left[-p_ip_{i+1}-\sqrt{1-p_i^2}\sqrt{1-p_{i+1}^2}\cos\left(\varphi_i-\varphi_{i+1}\right)+\frac{p_i^2}{2\gamma}-h\sqrt{1-p_i^2}\cos\varphi_i\right], \tag{3.3}$$

where $\gamma = J/(2D)$ and $h = g\mu_B H/(J\widetilde{S})$. This hamiltonian is a "classical counterpart" of (1.1), with the unusual correspondence of the quantum spin operators $\hat{\mathbf{S}}$ to classical unit vectors of length $\widetilde{S} = S+1/2$. For low values of the spin S this is, in our opinion, a sensible classical counterpart of (1.1), at difference with the usual *naïve* procedure [3,7].

In the present case the "skew" variational parameters $\sigma_k = 0$, and only the following three quantum renormalization parameters turn out to be relevant

$$\begin{aligned}
D_p &\equiv \langle\!\langle \eta_i^2 \rangle\!\rangle = \frac{\gamma\lambda}{N}\sum_k \frac{b_k}{2a_k}\left(\coth f_k - \frac{1}{f_k}\right) \quad , \\
D_\varphi &\equiv \langle\!\langle \xi_i^2 \rangle\!\rangle = \frac{\lambda}{N}\sum_k \frac{a_k}{2b_k}\left(\coth f_k - \frac{1}{f_k}\right) \quad , \\
D_{\delta\varphi} &\equiv \langle\!\langle \left(\xi_i - \xi_{i-1}\right)^2 \rangle\!\rangle = \frac{\lambda}{N}\sum_k 4\sin^2\frac{k}{2}\,\frac{a_k}{2b_k}\left(\coth f_k - \frac{1}{f_k}\right) \quad .
\end{aligned} \tag{3.4}$$

The usual field theoretic definition of the coupling constant is $\lambda = \left(\gamma\widetilde{S}^2\right)^{-1/2}$, and $f_k = \lambda a_k b_k/(2t)$, where $t = T/\left(J\widetilde{S}^2\right)$ is the dimensionless reduced temperature and

$$a_k^2 = 1+\gamma\left[\widetilde{h}-2\left(1-e^{-D_{\delta\varphi}/2}\right)+4\sin^2\frac{k}{2}\right], \quad b_k^2 = \delta^2\left[\widetilde{h}+4\tau\sin^2\frac{k}{2}\right] \quad . \tag{3.5}$$

Moreover, $\widetilde{h} = h\,e^{-D_\varphi/2}$, $\delta^2 = 1-D_p/2$ and $\tau = \delta^2\,e^{-D_{\delta\varphi}/2}$.

If we neglect, in view of the easy-plane character and according to the low-coupling approximation, terms of the order D_p^2 , the resulting effective hamiltonian can be written for classical spin variables $\mathbf{s}_i$ $\left(|\mathbf{s}_i|^2 = 1\ ,\ s_i^z = p_i/\delta\right)$ in the same form of (1.1), but for the appearance of a further anisotropy term in the exchange:

$$\begin{aligned}
\frac{\mathcal{H}_{\text{eff}}}{J\widetilde{S}^2} &= \delta^2\sum_{i=1}^N\left[-s_i^zs_{i+1}^z-\tau\left(s_i^xs_{i+1}^x+s_i^ys_{i+1}^y\right)+\frac{\left(s_i^z\right)^2}{2\gamma}-\widetilde{h}s_i^x\right]+t\sum_k\ln\frac{\sinh f_k}{f_k}+\Delta, \\
\Delta &= \frac{N}{2}\left[\widetilde{h}\left(D_\varphi+D_p-D_pD_\varphi/2\right)+e^{-D_{\delta\varphi}/2}\left(D_{\delta\varphi}+2D_p-D_pD_{\delta\varphi}\right)\right]+t\ln\delta\ .
\end{aligned} \tag{3.6}$$

If we let $\lambda \to 0$, keeping γ fixed (which corresponds to the limit $S \to \infty$), the Weyl symbol $\mathcal{H}$ is recovered.

The model parameters characteristic of $CsNiF_3$ correspond to the value of $\gamma = 1.3$. The low value of the spin $S = 1$ yields a rather high value of the coupling parameter, $\lambda = 0.58$, which is at the limit of reliability of the above framework. The evaluation of the thermodynamic quantities of $CsNiF_3$ has been done by the classical transfer matrix method, using the effective hamiltonian (3.6). The convergence of the method at lowest

temperatures has been checked and connected with the corresponding self-consistent gaussian approximation.

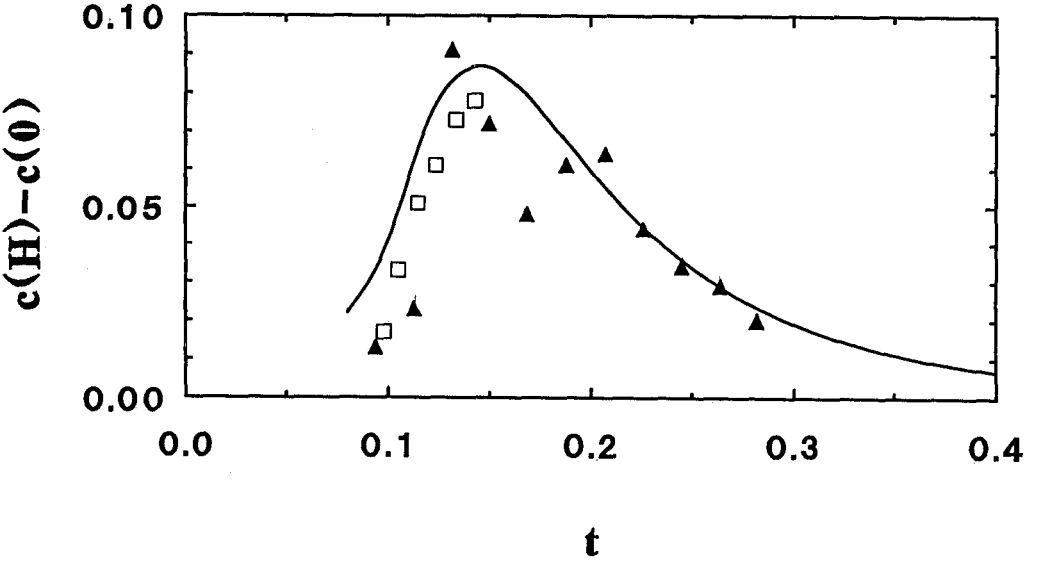

Figure 1 — *Excess specific heat per site (in units of k_B) versus the reduced temperature $t = T/(J\widetilde{S}^2)$. Model parameters characteristic of* $CsNiF_3$, *to which the values of $\gamma = 1.3$, $\lambda = 0.58$ do correspond. Field: $H = 5$ kG ($h = 0.022$). Solid line: quantum result. Squares: experimental data (ref. 4). Triangles: quantum Monte Carlo data (ref. 14).*

In the figures we report data for the *excess* specific heat per site, i.e. the difference between the specific heat and its value for zero Zeeman field. Indeed, the experimental measurements in $CsNiF_3$ made by Ramirez and Wolf [4] do report this difference, since it is not affected by the lattice contribution. In figure 1 the results of our calculation by use of the effective hamiltonian (3.6) are reported versus the reduced temperature $t = T/(J\widetilde{S}^2)$, and compared with the experimental data and with quantum Monte Carlo results by Wysin and Bishop [14]. The good agreement witnesses both the usefulness of the method and the correctness of the interaction parameters used for $CsNiF_3$.

In figure 2 the same results of the variational method are compared with the quantum results of the planar model and of the sine-Gordon model. Although the latter were calculated previously [10], we note that here we have used the correct procedure of approximating the *quantum* (Weyl ordered) hamiltonian to these two limits models, which gives rise to the same coupling parameter $\lambda = 0.58$ reported above. The quantum calculation has been performed within the framework of the variational method, using the corresponding effective potential. It is apparent that the anharmonicity of the in-plane exchange, which is retained by the planar model, is important, but is not enough to explain the full nonlinear contribution to the thermodynamics of the easy-plane ferromagnetic chain. The necessity of using a realistic model, which takes into account together out-of-plane fluctuations and anharmonic exchange is apparent.

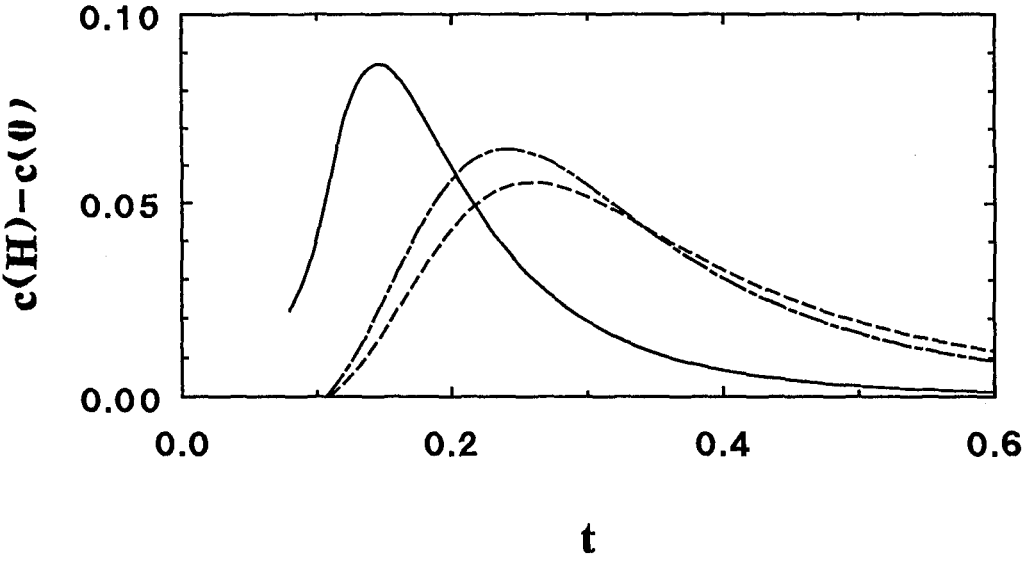

Figure 2 — *Quantum results for the excess specific heat. Units and model parameters as in figure* 1. *Dashed line: sine-Gordon approximation. Dash-dotted line: planar approximation. Solid line: full effective hamiltonian.*

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