Have Solitons Been Observed in $CsNiF_3$?

George Reiter
Physics Department, Brookhaven National Laboratory, Upton, New York 11973
(Received 30 June 1980)

It is shown that a noninteracting—spin-wave theory, in which the central component observed by neutron scattering is attributed to scattering from spin-wave—density fluctuations, gives intensities comparable to, or greater than, the intensities predicted by the soliton theory, and provides a better fit to the variation of the intensity with field, temperature, and wave vector. It is concluded that further work is needed to distinguish the soliton contribution to the central-peak intensity from the scattering by pairs of spin waves.

PACS numbers: 75.30.Ds

The central peak observed by neutron scattering from[1] $CsNiF_3$ in an external field has been interpreted in terms of the scattering from a dilute gas of solitons. This interpretation has been based upon a fit to a theoretical spectral function first derived by Kawasaki from heuristic arguments for the sine Gordon chain,[2] and subsequently argued by Mikeska[3] to be applicable to $CsNiF_3$ at temperatures well below the isotropic-anisotropic crossover. While this interpretation seemed to give a plausible explanation of the data, there remain some outstanding discrepancies. In particular, the observed intensity at large fields and low temperatures is much higher than the theoretical predictions.

I show here that the lowest-order harmonic spin-wave theory (which is nearly exact at low temperatures and/or high fields) provides a better description of the data than does the soliton theory. The central peak within the theory is due to the scattering from spin-wave—density fluctuations, and, if Mikeska's formula is believed, this mechanism provides intensity at least comparable to the scattering from solitons, over the range of the published data. In particular, the variation of the intensity with field, temperature, and wave vector is accurately reproduced over the range in which the harmonic theory can be expected to have some validity, which is most of the range of the data.

We take, for the Hamiltonian of $CsNiF_3$,

$$\mathcal{H}=\sum_i\left[-J\vec{S}_i\cdot\vec{S}_{i+1}+D(S_i^x)^2-g\mu_B H S_i^z\right], \qquad (1)$$

with $J=23.0\,^\circ\mathrm{K}$, $D=8.9\,^\circ\mathrm{K}$, $g=2.4$, and a quantum spin of 1. In the presence of a field at sufficiently low temperatures, the spins are nearly fully aligned, and one can use spin-wave theory to describe the system. The standard Holstein-Primakoff transcription to boson operators, taken to lowest order, i.e., $S_i^z=S-a_i^\dagger a_i$, $S_i^-=(S/2)^{1/2}a_i^\dagger$, $S_i^+=(S/2)^{1/2}a_i$, gives a quadratic Hamiltonian that is readily diagonalized. However, the $(S_i^x)^2$ term must be treated carefully.[4,5] What is required is that the boson equivalent gives the same matrix elements between states with one excitation and the vacuum for the operator $[(S/2)^{1/2}a^\dagger,(\tilde{S}^x)^2]$ as one obtains for $[S^-,(S^x)^2]$ between corresponding states. Here $(\tilde{S}^x)^2$ is the spin equivalent of $(S^x)^2$. This ensures that the spin-wave frequencies are correct to first order in D, for arbitrary S, and actually to second order, for $S=1$.[4,5] One finds then $(\tilde{S}_i^x)^2=\frac{1}{4}(2S-1)(a_i^\dagger+a_i)^2$ rather than $\frac{1}{2}S(a_i^\dagger+a_i)^2$. The result, for spin 1, is that the effective value of D is just half of what one would obtain from a naive substitution. Setting $\tilde{D}=D(S-\frac{1}{2})/S$ and $\tilde{H}=g\mu_B H$, we obtain the equivalent Bose Hamiltonian

$$\mathcal{H}=-N(J_0S^2+\tilde{H}S)+S\sum_q\left[(J_0+\tilde{D}+\tilde{H}-J_q)a_q^\dagger a_q+\tfrac{1}{2}\tilde{D}(a_q^\dagger a_{-q}^\dagger+a_qa_{-q})\right], \qquad (2)$$

where $J_q=2J\cos(qa)$ and a is the lattice parameter. The Hamiltonian is diagonalized by the transformation $a_q=\alpha_q b_q-\beta_q b_{-q}^\dagger$ and its conjugate, where $\alpha_q=\cosh\theta_q$, $\beta_q=\sinh\theta_q$, and $\tanh(2\theta_q)=\tilde{D}(J_0-J_q+\tilde{D}+\tilde{H})^{-1}$. Dropping the ground-state energy, we obtain

$$H=\sum_q\omega_q b_q^\dagger b_q, \quad \omega_q=S\{(J_0-J_q)^2+2(\tilde{D}+\tilde{H})(J_0-J_q)+2\tilde{D}\tilde{H}+\hat{H}^2\}^{1/2}. \qquad (3)$$

The correlation functions of interest are

$$\langle S_q^x(t)S_{-q}^x\rangle = \tfrac{1}{2}S(\alpha_q - \beta_q)^2[n_q \exp(i\omega_q t) + (n_q+1)\exp(-i\omega_q t)], \tag{4a}$$

$$\langle S_q^y(t)S_{-q}^y\rangle = \tfrac{1}{2}S(\alpha_q + \beta_q)^2[n_q \exp(i\omega_q t) + (n_q+1)\exp(-i\omega_q t)], \tag{4b}$$

$$\begin{aligned}\langle \delta S_q^z(t)\delta S_{-q}^z\rangle = N^{-1}\sum_{q_1,q_2} \Big[&(\alpha_{q_1}\alpha_{q_2}+\beta_{q_1}\beta_{q_2})^2 \exp[i(\omega_{q_1}-\omega_{q_2})t]n_{q_1}(n_{q_2}+1)\delta(q-q_1-q_2) \\ &+\tfrac{1}{2}(\alpha_{q_1}\beta_{q_2}+\beta_{q_1}\alpha_{q_2})^2\{\exp[i(\omega_{q_1}+\omega_{q_2})t]n_{q_1}n_{q_2} \\ &+\exp[-i(\omega_{q_1}+\omega_{q_2})t](n_{q_1}+1)(n_{q_2}+1)\}\delta(q-q_1-q_2)\Big],\end{aligned} \tag{4c}$$

where

$$\delta S_q^z = N^{-1/2}\sum_{q_1,q_2}[a_{q_1}^\dagger a_{q_2} - \langle a_{q_1}^\dagger a_{q_2}\rangle]\delta(q-q_1-q_2), \quad n_q = [\exp(\beta\omega_q)-1]^{-1}. \tag{5}$$

The magnetization is given by

$$\langle S_i^z\rangle = S - \langle a_i^\dagger a_i\rangle = S - N^{-1}\sum_q[(\alpha_q^2+\beta_q^2)n_q + \beta_q^2]. \tag{6}$$

The neutron-scattering cross section is proportional to $S_q^\alpha(\omega)$, where

$$S_q^\alpha(\omega) = (1/2\pi)\int_{-\infty}^{\infty} e^{i\omega t}\langle S_q^\alpha(t)S_{-q}^\alpha\rangle dt. \tag{7}$$

In Fig. 1, I show the longitudinal response function calculated numerically from (4c) for a set of parameter values $(q, \tilde{H}, T)$ characteristic of the experiments. The experiment actually measures some combination of the $S_q^\alpha(\omega)$ depending upon the orientation of the scattering vector. In the case that it lies along the chain, this is $S_q^z(\omega) + S_q^y(\omega)$. In the harmonic approximation, the transverse spectral function has no intensity near $\omega = 0$, and the central peak is contained entirely in the longitudinal response. Its intensity, which we take to be the initial value of the first term in (4c), vanishes at $T=0$, while the high-frequency band, corresponding to the neutron producing a pair of spin waves in the crystal, persists at $T=0$. The high-frequency band is currently the subject of experimental investigation, the results of which will be reported separately.[6]

Mikeska's formula for the contribution to $S_q(\omega)$ due to solitons, as modified by Steiner[7] and by Leung and Huber[8] to include the transverse contribution, is

$$S_q^{\text{sol}}(\omega) = \frac{64}{\pi}\frac{1}{KTqc}\exp\left(-\frac{8mJa}{KT}\right)\exp\left(-\frac{4mJa}{KT}\frac{\omega^2}{c^2q^2}\right)\left(\frac{\pi q}{2m}\right)^2\left\{\left[\sinh\left(\frac{\pi q}{2m}\right)\right]^{-2}+\left[\cosh\left(\frac{\pi q}{2m}\right)\right]^{-2}\right\}, \tag{8}$$

where $m = (H/Ja^2)^{1/2}$ and $c = aS(2JD)^{1/2}$. The result in the literature[3,7,8] differs from Eq. (8) by a factor of 2π, because of a different definition of $S_q(\omega)$. Equations (7) and (8) are consistent, and (7) is the usual definition. The formula is only valid for nonrelativistic particles, but the relativistic corrections[8] are not large enough to affect our argument, and (8) has the virtue that the integrated intensity can be simply calculated.

In Fig. 2, I show the integrated intensity in the central peak as calculated from (4c) and (8). The relative intensity of the two spin-wave and soliton contributions is shown correctly, while the experimental intensity has been normalized to agree with the spin-wave result at the lowest temperature. In Fig. 3, I show the same comparison with the field being varied, the experimental results being normalized at the highest field. In Fig. 4, I show the intensity as a function of wave vector for a fixed field and temperature, compared with the experimental results. A similarly good fit is obtained from Eq. (8) if

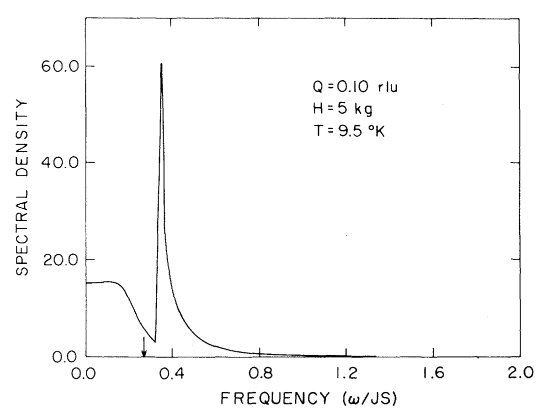

FIG. 1. The longitudinal spectral density in $CsNiF_3$. The arrow locates the spin-wave frequency.

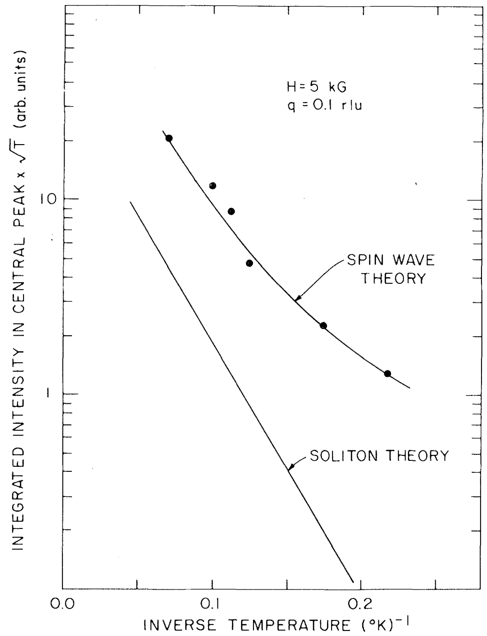

FIG. 2. Variation of the intensity in the central peak with temperature. The experimental points are the data of Kjems and Steiner (Ref. 1).

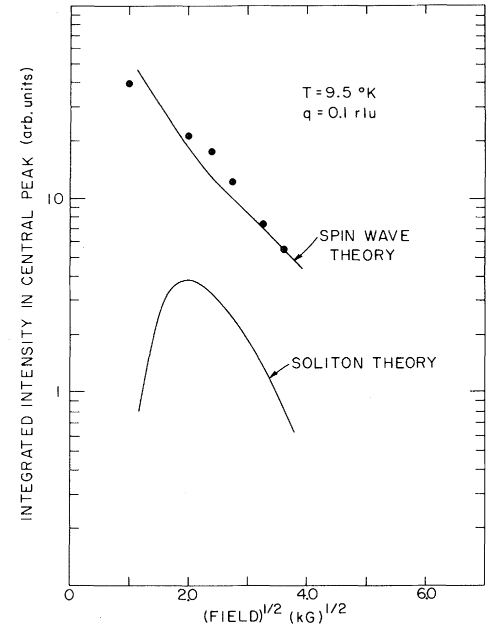

FIG. 3. Variation of the intensity in the central peak with field. The effect of the variation of the soliton shape with field, which is responsible for the downturn of the theoretical curve, was omitted in Ref. 1 and accounts for the difference between this figure and Fig. 2 of Ref. 1.

m is allowed to be an adjustable parameter, but the m required corresponds to a field of roughly four times the actual applied field.[9] There are, of course, no adjustable parameters in the spin-wave theory.

The spin-wave theory is rigorously correct in the limit of high fields, and nearly so for arbitrary fields as $T \to 0$. Some idea of its validity at the fields and temperatures of the experiments can be had by comparing the prediction of the magnetization in the classical limit with the exact results from transfer-matrix calculations.[10] We find that for fields above 5 kG and temperatures below 10 °K, the deviation of the spin-wave result from the exact result is at most 10% of the value of the magnetization.

The spin-wave theory makes one prediction that is not borne out by the experiments. The central-peak linewidth from either theory is proportional to q, but the magnitude of the linewidth predicted by the spin-wave theory is about twice the maximum experimental value for a given q and is nearly independent of temperature, whereas the experiments do show a significant temperature variation.[1]

The most charitable interpretation of the data is that they consist of the sum of a spin-wave and soliton contribution, of comparable intensities at low fields and/or high temperatures. To show unambiguously that there was a soliton contribution would therefore require knowing the theoretical spin-wave contribution, to an accuracy of better than, say, 50%, and comparing with absolute intensity data, obtained for instance by normalizing to the low-temperature, single-spin-wave intensity. This requires that a perturbative

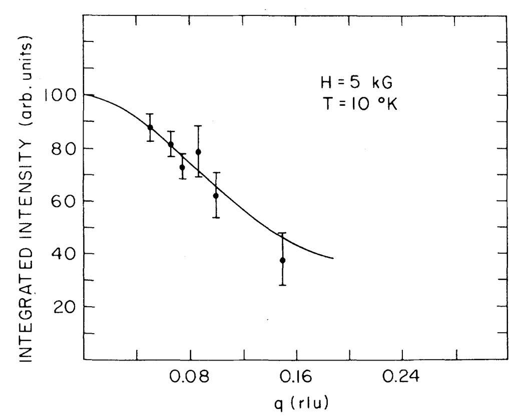

FIG. 4. Variation of the intensity of the central peak with wave vector. The solid line is the spin-wave prediction.

treatment of the spin-wave theory, taking into account anharmonic effects, be done with the sufficient rigor that its accuracy could be assessed at the temperatures and fields of the experiment. A subtraction could then be done to determine the amplitude of the nonperturbative effects. We expect that the contribution of breather modes, which are essentially bound pairs of spin waves, would be included in the perturbation theory, and would not make any separate contribution to the intensities, although they might produce observable effects in the shape of the spectrum. The nonperturbative contribution to the total intensity could presumably be associated with the solitons, and may be describable by (8). Inasmuch as no such separation has been accomplished to date, one cannot say that solitons have been observed in $CsNiF_3$.

I would like to acknowledge valuable conversations with J. Kjems, M. Steiner, J. Loveluck, and H. J. Mikeska. I would also like to thank H. J. Mikeska for pointing out an error in the relative normalization of the spin-wave and soliton contributions, and Nancy White for her patience and skill in preparing several versions of this manuscript. This research was supported by Division of Basic Energy Sciences, U. S. Department of Energy, under Contract No. DE-AC02-76CH00016.

[1] J. K. Kjems and M. Steiner, Phys. Rev. Lett. 41, 1137 (1978).
[2] K. Kawasaki, Prog. Theor. Phys. 55, 2029 (1976).
[3] H. J. Mikeska, J. Phys. C 11, L29 (1978).
[4] G. Reiter, Ph.D. thesis, Stanford University (unpublished).
[5] P.-A. Lindgard and A. Kowalska, J. Phys. C 9, 2081 (1976).
[6] G. Shirane, private communication.
[7] M. Steiner, J. Appl. Phys. 50, 7395 (1979).
[8] K. M. Leung and D. L. Huber, Solid State Commun. 32, 127 (1979).
[9] M. Steiner, J. Appl. Phys. 50, 7395 (1979).
[10] T. Schneider, E. Stoll, and H. R. Jauslin, to be published.

Substitutional Donors and Core Excitons in Many-Valley Semiconductors

M. Altarelli
Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 28 July 1980)

The shallow-deep instability of substitutional donors and core excitons is discussed, with inclusion of all intervalley interactions. Shallow levels result in Si for a screened point-charge potential, because intervalley overlap and kinetic energy balance the potential-energy terms, which are severely reduced, at substitutional sites, by umklapp effects. Contrary to recent claims based on consideration of potential energy only, many-valley interactions cannot therefore be invoked to predict deep core-exciton levels in Si.

PACS numbers: 71.55.Fr, 71.35.+z

Much interest in impurity states in semiconductors with many-valley bands has recently been stimulated by the experimental discovery that interstitial muonium is a deep donor in Si and Ge,[1] by suggestions that core-exciton levels may also be deep,[2] and by the theoretical discovery that drastic modifications of the traditional[3] effective-mass approach are needed to account for intervalley interactions properly.[4-7] In the theoretical description of muonium in Si and Ge,[8,9] two effects have been considered to explain its deep-level character. One is the point-charge nature of the muon potential, as contrasted with weaker pseudopotentials for impurity atoms with core electrons; the other is the location of the muon at the interstitial site, where intervalley matrix elements of the point-charge potential are ~3–4 times larger than at the substitutional site.[9] Very recently,[10] it has been argued that the second effect (the site dependence) plays no role and that, in fact, deep levels result in Si from a screened point-charge potential, irrespective of its location. In view of the relevance of the question in general, and in particular for the problem of core excitons, which can be described as electrons bound by the nearly pointlike charge of a core hole in a host atom, the purpose of the present Letter is to discuss the energy levels of a

