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Soliton contribution to the electron paramagnetic resonance linewidth in a two-dimensional antiferromagnet with a staggered field

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Abstract
In this paper we study the interactions between magnons and a soliton in a classical and isotropic two-dimensional Heisenberg antiferromagnet in the presence of a staggered field applied perpendicularly to the XY plane. The temperature dependence of the linewidth is calculated using the dynamic spin correlation function derived from soliton–magnon scattering.

The study of the interaction between magnons and topological excitations in classical magnetic systems is of fundamental importance, as it is the starting point for a thermodynamic analysis of the system, as shown by Currie *et al* [1]. As pointed out by Zaspel *et al* [2] it is also important in the study of the dynamics of vortices. As is well known, topological excitations contribute to a central peak in the dynamical relaxation function and this peak is hard to detect in an unambiguous way. Nevertheless, the signature of the topological excitations can be seen in the electron paramagnetic resonance (EPR) linewidth [2]. In this paper we will focus on these fluctuations as observed through EPR line broadening, which occurs in a narrow temperature range just above the Néel temperature T_N, in the nearly 2D antiferromagnetic case. Waldner [3] showed experimentally that classical layered antiferromagnets exhibited an Arrhenius EPR linewidth given by $\Delta H \sim \exp(E_s/T)$, where E_s is the soliton energy, and T is the temperature dependence immediately above T_N. In [3, 4] it was shown that the measured E_s energy in the EPR linewidth temperature dependence agreed with the energy of the Belavin and Polyakov [5] soliton to within a few per cent for four different compounds, and it was implied that solitons contributed to the EPR linewidth in the critical fluctuation region.

The EPR linewidth is related to the time-dependent spin correlation function, and consequently static solitons cannot contribute to the linewidth. To interpret the observed Arrhenius behaviour it is necessary to calculate the dynamic soliton contribution to the EPR

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linewidth or equivalently the time-dependent correlation function in the critical fluctuation region. Gouvea *et al* [6], through the results of the interaction between topological excitations and spin waves, showed a rise to the central peak in the frequency-dependent correlation function. Zaspel [7] showed that soliton motion also results in a central peak. Another important contribution to soliton dynamics is the interaction of spin waves with the soliton; consequently this interaction contributes to time-dependent spin correlation functions.

Solitons interacting with magnons have been studied in two-dimensional nonlinear sigma models (isotropic [8] and anisotropic [9]) and in two-dimensional anisotropic ferromagnets [10]. It has been found that the quantum corrections to the classical soliton, or vortex energy, given by the zero-point energy of the spin waves measured with respect to the vacuum can change strongly the classical picture, introducing interactions between solitons [8] as well as an internal degree of freedom [9].

Our purpose in this paper is to study the EPR linewidth due to interaction between spin waves and solitons, in a two-dimensional antiferromagnet with a uniform staggered field applied perpendicularly to the plane. Asano *et al* [11] reported electron spin resonance of the $S = 1/2$ antiferromagnet Heisenberg chain, Cu purimidine. The effect of the staggered field was clearly observed for the ESR linewidth. A systematic study of coupled $S = 1/2$ antiferromagnet chains in an effective staggered field was performed by [12]. The mechanisms generating the staggered fields in real magnets was discussed in [13–16]. Recently many works have been published focusing on the importance of this study [17, 18]. All materials studied so far are highly one dimensional or quasi-one dimensional. As far as we know there are at the present time no experimental results for an antiferromagnet in 2D in the presence of a staggered field. Although there is a lot of theoretical work dedicated to the staggered field in one-dimensional antiferromagnets, the number of papers relating to 2D models is small [19, 20]. We found that the lowest-order effect of an inhomogeneous soliton is to produce an elastic scattering centre for the spin waves, and we obtained the solution for the EPR linewidth.

We start by considering the model described by the following Hamiltonian:

$$H = \sum_{\langle ij\rangle}[J\mathbf{S}_{i,j}\cdot(\mathbf{S}_{i+1,j}+\mathbf{S}_{i,j+1}) + g_0\mu_0\mathbf{B}\cdot(-1)^i\mathbf{S}_{i,j}], \tag{1}$$

where the summation extends over all sites of a square lattice, J is the positive exchange constant, $\mathbf{S}_{i,j}$ is the spin vector at site (i, j), g_0 is the gyromagnetic ratio, $\mu_0 = e/2mc$ is the Bohr magneton divided by the Planck constant and $\mathbf{B}$ is the magnetic field which will be taken to point in the third direction $\mathbf{B} = B\hat{z}$. The antiferromagnetic model in a staggered field is a very convenient model to study, since at low temperature it can be mapped in the nonlinear sigma model [21, 22], with the staggered field acting as a source. Then, the Hamiltonian can be written as

$$H = \frac{J}{2}\int[(\partial_0\mathbf{l}_n)^2 - (\partial_\alpha\mathbf{l}_n)^2 + 2hl_3]\,\mathrm{d}^2x \qquad \alpha = 1, 2, \tag{2}$$

where $h = g_0\mu_0\mathbf{B}/(4JS)$. It is useful to resolve the constraint $\mathbf{l}_n^2 = 1$ explicitly using the spherical parameterization $\mathbf{l}_n = S(\sin\theta\cos\phi, \sin\theta\sin\phi, \cos\theta)$ in terms of which

$$H = \frac{J}{2}\int\left[\frac{1}{c^2}\left(\frac{\partial\theta}{\partial t}\right)^2 - (\vec{\nabla}\theta)^2 + \sin^2\theta\left[\frac{1}{c^2}\left(\frac{\partial\phi}{\partial t}\right)^2 - (\vec{\nabla}\phi)^2\right] + 2h\cos\theta\right]\mathrm{d}^2x, \tag{3}$$

where $c = 2aJS$ is the spin-wave velocity. The parameter a is the lattice spacing. The equations of motion following from equation (3) are:

$$\nabla^2\theta - \frac{1}{c^2}\frac{\partial^2\theta}{\partial t^2} = \sin\theta\cos\theta\left[(\vec{\nabla}\phi)^2 - \frac{1}{c^2}\left(\frac{\partial\phi}{\partial t}\right)^2\right] + h\sin\theta, \tag{4}$$

$$\nabla^2\phi - \frac{1}{c^2}\frac{\partial^2\phi}{\partial t^2} = -2\cot\theta\left[(\vec{\nabla}\theta\cdot\vec{\nabla}\phi) - \frac{1}{c^2}\frac{\partial\phi}{\partial t}\frac{\partial\theta}{\partial t}\right]. \tag{5}$$

The static solution ϕ_s for equations (4) and (5) is $\phi_s = q\arctan(y/x)$, were the parameter $q = 0, 1, 2, \ldots$ plays the role of the topological charge of the soliton. We can write the localized solutions of equations (4) and (5) in polar coordinates in the form

$$\theta = \theta_s(r), \qquad \theta_s(0) = 0, \qquad \theta_s(\infty) = 2\pi, \qquad \phi_s(\varphi, t) = q\varphi - \Omega t. \tag{6}$$

Here, Ω is the internal precession frequency of the soliton and can be determinate, through the number of bound magnons N. Substituting equation (6) into equations (4) and (5), we notice that equation (5) is automatically satisfied. Then, from equation (4), we obtain

$$\frac{1}{k_0^2}\left(\frac{d^2\theta_s}{dr^2} + \frac{1}{r}\frac{d\theta_s}{dr}\right) + \left(1 - \frac{q^2}{k_0^2 r^2}\right)\sin\theta_s\cos\theta_s - \frac{h}{k_0^2}\sin\theta_s = 0, \tag{7}$$

where $k_0^2 = \Omega^2/c^2$. By convenience we have introduced $l_0^2 = 1/k_0^2$ and $H = h/k_0^2$. The solution of equation (7) was encountered by Kosevich *et al* [23]. The magnetization in equilibrium is $[\theta_s(r) = \theta_0]$ from the soliton ($r \to \infty$), and $\theta_s(r) = 0$ for $r = 0$. It follows from the latter condition that, as $r \to 0$

$$\theta_s(r) = (r/r_0)^{|q|}, \qquad r_0 = \text{constant}. \tag{8}$$

Notice that $\theta_s(r)$ does not depend of H; however, the behaviour for $r \to \infty$ depends on H. For $H \neq 0$ and 1 the solution has the following behaviour at infinity:

$$\theta_s(r) = \theta_0 - \frac{q^2 H^2}{\sqrt{1 - H^2}}\left(\frac{l_0}{r}\right)^2 \qquad H \neq 0 \text{ and } 1. \tag{9}$$

The main macroscopic characteristic of the soliton is its energy. It is well known that soliton energy in an infinite crystal diverges logarithmically. Hence, with a logarithmic accuracy the soliton energy is

$$E_s = \pi J q^2(1 - H^2) a M_0^2 \ln[R A(H)/l_0] \tag{10}$$

where a is the lattice parameter, M_0 is the z-magnetization and R is a cut-off soliton radius. The function $A(H)$ is a finite term with $R \to \infty$. The function $A(H)$ can be found by numerical methods. As was shown by Kosevich *et al* [23], this function varies between 0.2 for ($H = 1$) and 4.2 for ($H = 0$). More details about this solution can be found in [20]. This solution has been criticized in a comment made by Sheka [24], where it is affirmed that the energy of the solution diverges with the size of the soliton, as in fact it diverges. He concludes that a solution for soliton precessional should not exist. However, we have two solution types here: one in which the soliton is considered not to be precessional—in this case it is sufficient to take $\Omega = 0$. This solution was shown in [25], and was not questioned. The fact is that the solution with $\Omega = 0$ presents a finite energy. The other solution, $\Omega \neq 0$, could be applied to finite systems such as nano-discs, where the energy is finite. The fact is that the solution obtained by Kosevich [23] for equation (7) really showed a divergence that is logarithmic in the energy. Nevertheless, we believe that the arguments mentioned above reinforce the utilization of solutions (8) and (9).

In order to determine the behaviour of magnons in the presence of a soliton, we assume that the spin polar angle is given by $\theta(\vec{r}, t) = \theta_s(r) + \eta(\vec{r}, t)$, and the spin azimuthal angle by $\phi = \vec{k}\cdot\vec{r} - \omega_\phi t$. Here, $\eta(\vec{r}, t)$ are assumed to be a small quantity, i.e. $\eta(\vec{r}, t) \ll 1$, which reduce to the magnon solutions if no solitons are present. In the presence of a soliton, $\eta(\vec{r}, t)$ gives the change in the soliton configuration as a result of the soliton–magnon interaction. Considering that the asymptotic component is $\theta_s(r) = 0$ (equations (6), (8) and (9)), since $\theta_0 \ll 1$, we can substitute $\theta(\vec{r}, t)$ in equation (4), neglecting quadratics terms in $\eta(\vec{r}, t)$, and obtain the equation of motion for magnons in the presence of a soliton as

$$\nabla^2\eta - \frac{1}{c^2}\frac{\partial^2\eta}{\partial t^2} = \eta\left[(\vec{\nabla}\phi_s)^2 - \frac{1}{c^2}\left(\frac{\partial\phi_s}{\partial t}\right)^2 + h\right]. \tag{11}$$

The solutions for equation (12) represent the out-of-plane spin waves. We showed the solution for equation (12) in [20]; it can be written as

$$\eta(\vec{r},t) = C_0 J_\mu(kr)\mathrm{e}^{-\mathrm{i}[n\varphi+\omega_\theta t]}, \tag{12}$$

where $J_\mu(kr)$ is the Bessel function, $\mu = \sqrt{n^2+q^2}$, and $n = 0, 1, 2, 3, \ldots$ represent the quantum number of angular momentum for the out-of-plane spin waves. The parameter k is the respective wavevector. The constant C_0 is determined through the normalization of the eigenfunctions. The equation (11) admits the dispersion relation $\omega_\theta^2 = k^2c^2 - \Omega^2 + hc^2$. Sheka, in the comment [24], criticized the choice of this solution of plane waves in the form $\eta(\vec{r},t) = \exp(\mathrm{i}[\vec{k}\cdot\vec{r} - \omega t])$, in which it is not a correct mathematical object. We agree; however, in the asymptotic limit the Bessel function can be written as a linear combination of the Hankel functions of first and second type, where they have form of plane waves. Therefore, we have used the solution $\eta(\vec{r},t) = \exp(\mathrm{i}[\vec{k}\cdot\vec{r} - \omega t])$ as an artifice, just to calculate the dispersion relation. The solution equation (12) to equation (11) is correct, and can be verified easily by the reader.

To calculate the out-of-plane spin-correlation function we use the soliton structure factor

$$f^i(\vec{k},t) = \int l^i(\vec{r},t)\mathrm{e}^{\mathrm{i}\vec{k}\cdot\vec{r}}\,\mathrm{d}^2r, \tag{13}$$

where $i = x, y$ is the i component of the sublattice magnetization with time dependence resulting from the soliton–magnon interaction. This structure factor contains a static contribution from the static solution and a time-dependent contribution from the soliton–magnon scattering. For $\eta \ll 1$ and $\theta_\mathrm{s} \ll 1$ we made the approximation $\sin[\theta_\mathrm{s}+\eta] \approx \theta_\mathrm{s}+\eta$. The term $\vec{k}\cdot\vec{r}$ in equation (13) can be written in the form $\vec{k}\cdot\vec{r} = |\vec{k}||\vec{r}|\cos(\alpha_k - \varphi)$, where α_k is the angle of the vector $\vec{k}$ with the x axis, and φ is the angle of the vector $\vec{r}$ with the x axis. For simplicity we will make $\alpha_k = 0$. In this paper, we will calculate the EPR linewidth for the ground state ($n = 0$) and a soliton with topological charge $q = 1$, because these values describe the state of lower energy. Thus, we can write the structure factor as:

$$f^{x,y}(\vec{k},t) = \left[\int_0^R J_1(kr)\mathrm{e}^{-\mathrm{i}\omega_\theta t} + \int_0^d \theta_{s_{r\to 0}}(r) + \int_d^\infty \theta_{s_{r\to\infty}}\right] \times \left\{\begin{array}{c}\cos(\varphi + kr\cos\varphi - \omega_\phi t)\\ \sin(\varphi + kr\cos\varphi - \omega_\phi t)\end{array}\right\} \mathrm{e}^{\mathrm{i}kr\cos(\varphi)} r\,\mathrm{d}r\,\mathrm{d}\varphi. \tag{14}$$

Here, $\theta_{s_{r\to 0}}$ and $\theta_{s_{r\to\infty}}$ are equations (8) and (9), respectively. We remark that in this equation f^x and f^y corresponds to cos and sin, respectively. In the first integral R is a cut-off. The parameter d in the limits of integration is a number for which equations (8) and (9) have the same value. From equations (8) and (9) we obtain

$$d = \frac{\sqrt[3]{r_0 H^2 l_0^2}}{(1-H^2)^{1/6}}. \tag{15}$$

Integrating the equation (14), we obtain:

$$f^x_{(1)}(k,t) = \frac{\mathrm{i}R\pi\mathrm{e}^{-\mathrm{i}(\omega_\theta+\omega_\phi t)}}{3k} W_1(kR) + \frac{\mathrm{i}\pi\mathrm{e}^{-\mathrm{i}\omega_\phi t}d^2}{2r_0} W_2(kd) - \frac{\mathrm{i}H^2 l_0^2\mathrm{e}^{-\mathrm{i}\omega_\phi t}}{\sqrt{1-H^2}} W_3(kd), \tag{16a}$$

$$f^y_{(1)}(k,t) = \frac{R\pi\mathrm{e}^{-\mathrm{i}(\omega_\theta+\omega_\phi t)}}{3k} W_1(kR) + \frac{\pi\mathrm{e}^{-\mathrm{i}\omega_\phi t}d^2}{2r_0} W_2(kd) - \frac{H^2 l_0^2\mathrm{e}^{-\mathrm{i}\omega_\phi t}}{\sqrt{1-H^2}} W_3(kd), \tag{16b}$$

where

$$W_1(kR) \equiv [J_0(kR)J_1(2kR) - 2J_0(2kR)J_1(kR)], \tag{17a}$$

$$W_2(kd) \equiv \frac{J_2(2kd)}{k}, \tag{17b}$$

$$W_3(kd) \equiv -kdF[(1/2);(3/2,2);-(kd)^2]+1. \tag{17c}$$

The function $F[(1/2);(3/2,2);-(kd/2)^2]$ is a hypergeometric confluent function. In the limit $k \to 0$, the equations (16) are:

$$f^x_{(0)}(t) = -\frac{\mathrm{i}\pi H^2 l_0^2 \mathrm{e}^{-\mathrm{i}\omega_\phi t}}{\sqrt{1-H^2}}, \tag{18a}$$

$$f^y_{(0)}(t) = -\frac{\pi H^2 l_0^2 \mathrm{e}^{-\mathrm{i}\omega_\phi t}}{\sqrt{1-H^2}}. \tag{18b}$$

The EPR linewidth is the temporal integral of the four-spin-correlation function and is given by [2, 26, 27, 32]

$$\Gamma = \frac{k_\mathrm{B}T}{2\chi_\perp \hbar^2}\sum_{k,k'} A(k_0)\,\mathrm{Re}\int \mathrm{e}^{-\mathrm{i}\omega_r t}\langle l^i_k(t) l^i_{-k'}(t) l^i_{-k}(0) l^i_{k'}(0)\rangle\,\mathrm{d}t, \tag{19}$$

where $\chi_\perp$ is the uniform susceptibility, ω_r is the resonance frequency and $A(k_0)$ is related to the Fourier coefficients of the dipolar interaction evaluated at the antiferromagnetic wavevector $(k_0 = \pi/a)$.

Considering only incoherent scattering from independent solitons [28] at different centres $\mathbf{r}_j$, then $l^i(\vec{r},t) = \sum_j l(\mathbf{r}-\mathbf{r}_j,t)$, and the sums over the pair centres yield [29] $n\langle f(k,t)f(-k,t)f(k,0)f(k,0)\rangle$ for the correlation function, where $f = (f^x + f^y)/2$ is a symmetrized structure factor [28] and n is the soliton density. Thus, the leading soliton contribution to the linewidth is [2]

$$\Gamma \sim \frac{nk_\mathrm{B}T}{\chi_\perp}\frac{S^4}{2\pi}\sum_k^2 \mathrm{Re}\left[\int_0^\infty \langle f^2_{(0)}\rangle\langle f_{(0)} f_{(1)}\rangle \mathrm{e}^{\mathrm{i}4\omega_\phi t}\mathrm{e}^{\mathrm{i}\omega_r t}\,\mathrm{d}t\right], \tag{20}$$

with $f_{(0)} = (f^x_{(0)} + f^y_{(0)})/2$, $f_{(1)} = (f^x_{(1)} + f^y_{(1)})/2$; assuming that magnetization relaxes by diffusion the complex magnon frequency can be written as $\omega_\phi = ck + \mathrm{i}Dk^2/2$, where D is the diffusion coefficient. In equation (20), because of the existence of temporal symmetry, we made an exchange in the sign of the exponentials to facilitate the results. The soliton density is proportional to $\mathrm{e}^{-E_\mathrm{s}/2T}$, where E_s is the soliton energy. To a first approximation we can use the diffusion coefficient obtained through dynamic scaling by Chakravarty *et al* [30], $D = \xi\sqrt{T/\chi_\perp}$, and the correlation length from Takahashi's [31] modified spin wave theory for 2D AFM, $\xi = (1/8\sqrt{2}\mathrm{e}^{\pi/2})\mathrm{e}^{E_\mathrm{s}/2T}$.

Performing the integral, we obtain:

$$\Gamma \sim \frac{nk_\mathrm{B}T}{\chi_\perp}\frac{S^4\pi^4H^6l_0^6}{2\pi\sqrt{1-H^2}}\sum_k^2\left[\frac{DRk^2}{24(1-H^2)}\frac{W_1(kR)}{(\omega_r+\omega_\theta+4ck)^2+4D^2k^4}\right.$$
$$\left. - \frac{Dk^2d^2}{16r_0(1-H^2)}\frac{W_2(kd)}{(\omega_r+4ck)^2+4D^2k^4} + \frac{Dk^2H^2l_0^2}{8(1-H^2)^4}\frac{W_3(kR)}{(\omega_r+4ck)^2+4D^2k^4}\right]. \tag{21}$$

The sum in equation (21) can be converted to an integral in two dimensions. Since well-defined spins waves exist is $ka > 1$, the lower limit of the k integral is $1/a$. We integrated the sum in k numerically. The solution EPR linewidth can be shown in graphical form.

In figure 1, we have the behaviour of the EPR linewidth as a function of the temperature, for a staggered field equal to $0.2J$. Here, we took $J = k_\mathrm{B} = \hbar = 1$. The behaviour of the EPR linewidth is very similar, to that presented in the literature. In figure 2, we have the

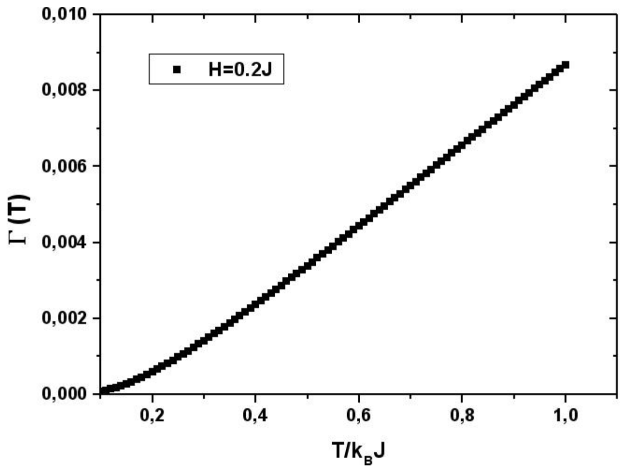

Figure 1. Graph of the EPR linewidth as a function of temperature for $H = 0.2J$, to due soliton–magnon interaction.

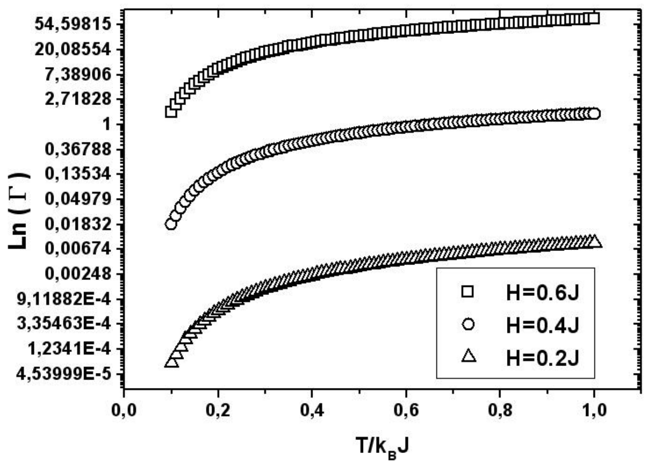

Figure 2. Graph of the logarithm of the EPR linewidth for $H = 0.2J$, $H = 0.4J$ and $H = 0.6J$.

behaviour of the logarithm of the EPR linewidth as a function of the temperature. In this figure, we can see an increase in the EPR linewidth with the increase in the staggered field. Our theoretical calculation in equation (21) can be compared directly with experimental data for the EPR linewidth once the experimental data for a 2D antiferromagnet are available.

In conclusion we have calculated the EPR linewidth in two-dimensional antiferromagnets in the presence of a staggered field. The measurements of the EPR linewidth provide an indirect method for experimental detection of solitons. The compounds R_2BaNiO_5 and Cu benzoate, for instance, can be treated as a one-dimensional antiferromagnet model immersed in a strong effective staggered field. In summary, solitons interacting with magnons in the classical 2D AFM result in an EPR linewidth with a dominant $\exp(E_s/T)$ temperature dependence. This dependence can occur only if solitons are present in the fluctuation region.

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