Nuclear Magnetism and Electronic Order in ^{13}C Nanotubes

Bernd Braunecker,[1] Pascal Simon,[1,2,3] and Daniel Loss[1]

[1]*Department of Physics, University of Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland*
[2]*Laboratoire de Physique et Modélisation des Milieux Condensés, CNRS and Université Joseph Fourier, BP 166, 38042 Grenoble, France*
[3]*Laboratoire de Physique des Solides, CNRS UMR-8502, Université Paris Sud, 91405 Orsay Cedex, France*

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Single wall carbon nanotubes grown entirely from ^{13}C form an ideal system to study the effect of electron interaction on nuclear magnetism in one dimension. If the electrons are in the metallic, Luttinger liquid regime, we show that even a very weak hyperfine coupling to the ^{13}C nuclear spins has a striking effect: The system is driven into an ordered phase, which combines electron and nuclear degrees of freedom, and which persists up into the millikelvin range. In this phase the conductance is reduced by a universal factor of 2, allowing for detection by standard transport experiments.

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The physics of conduction electrons interacting with localized magnetic moments is central for numerous fields in condensed matter such as nuclear magnetism [1], heavy fermions [2], or ferromagnetic semiconductors [3–6]. Nuclear spins embedded in metals offer an ideal platform to study the interplay between strong electron correlations and magnetism of localized moments in the RKKY regime. In two dimensions the magnetic properties of the localized moments [7,8] depend indeed crucially on electron-electron interactions [9–13]. In one-dimensional (1D) systems such as single wall carbon nanotubes (SWNTs) electron correlations are even more important. For metallic (armchair) SWNT they lead to Luttinger liquid physics [14–16]. Recently, SWNTs made of ^{13}C, forming a nuclear spin lattice, have become experimentally available [17–20]. Motivated by this we study here nuclear magnetism in metallic ^{13}C SWNTs. We show that even a weak hyperfine interaction can lead to a helical magnetic order of the nuclear spins (see Fig. 1) coexisting with an electron density order that combines charge and spin degrees of freedom. The ordered phases stabilize each other, and the critical temperature undergoes a dramatic renormalization up into the millikelvin range due to electron-electron interactions. In this new phase the electron spin susceptibility becomes anisotropic and the conductance of the SWNT drops by a universal factor of 2.

The drastic restructuring of the electron wave functions through the renormalization is very different from the case of two [7,8] or three dimensions [1] where it is, in comparison, weak. The same renormalization leads to considerable anisotropy in the electron system: The nuclear magnetic field spontaneously breaks the spin rotational symmetry; it rotates in a plane, which we can associate with the spin (x, y) directions (see Fig. 1). This plane is singled out as an easy plane through the stabilization of the electron density wave, and electron correlation functions become anisotropic between the spin (x, y) plane and the spin z direction. We illustrate this behavior below through the calculation of the electron spin susceptibilities. We emphasize that this anisotropy is a crucial feature of the SWNT system studied here and appears *spontaneously* due to strong renormalization of the RKKY interactions. This distinguishes our system, in particular, from models with built-in easy-axis anisotropy [21].

Model.—We assume that the electrons are confined in a single mode $\psi_\perp$ in the directions perpendicular to the tube axis. The nuclear spins $I = 1/2$ of the ^{13}C ions on a circular cross section have identical overlaps with this transverse mode, and so identical couplings to the electrons. Through their indirect RKKY interaction over the electron gas they are therefore locked in a ferromagnetic alignment (see Fig. 1). This RKKY interaction, described below, overrules furthermore the direct dipolar interaction between the nuclear spins. The latter is very small [22], $\sim 10^{-11}$ eV, and shall be neglected henceforth. This allows us to treat the nuclear spins as a 1D chain of *large* $\tilde{I} = IN_\perp$ spins, composed of the sum of the $N_\perp \sim 50$ spins around a circular cross section. Because of this, Kondo physics, which requires small quantum spins, can be excluded from the beginning.

Hence, we model the SWNT by a 1D nuclear spin lattice of length L coupled through the hyperfine interaction to a 1D electron gas. The Hamiltonian resembles that of a Kondo lattice $H = H_{\rm el} + A\sum_i \hat{\mathbf{S}}_i \cdot \hat{\mathbf{I}}_i$, where i runs over the 1D lattice sites with positions r_i, $\hat{\mathbf{I}}_i = (\hat{I}_i^x, \hat{I}_i^y, \hat{I}_i^z)$ is the effective nuclear spin of size $\tilde{I} = IN_\perp$, $\hat{\mathbf{S}}_i = (\hat{S}_i^x, \hat{S}_i^y, \hat{S}_i^z)$ is the electron spin operator at site i, and $A = A_0/N_\perp$ is the on-site hyperfine interaction constant A_0 weighted by the transverse electron mode. In contrast to the usual Kondo-lattice model, $H_{\rm el}$ describes the *interacting* electrons and is defined in Eq. (2) below.

The precise value of A_0 in SWNTs is unknown. Estimates in the literature [23] provide values of $A_0 \sim 10^{-7}$–10^{-6} eV, depending much on the curvature of the

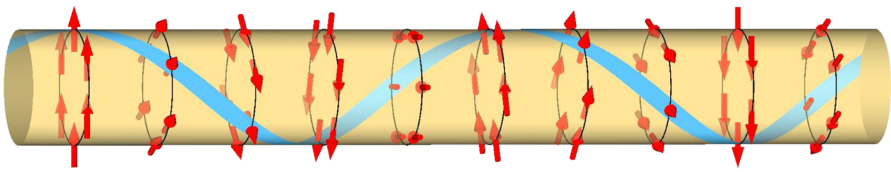

FIG. 1 (color online). Illustration of the helical nuclear magnetism (indicated by the blue ribbon) of the single wall ^{13}C nanotube (SWNT), which emerges below a critical temperature through strong renormalization of the hyperfine coupling by electron correlations. The nuclear spins (red arrows) order ferromagnetically on a cross section of the SWNT and rotate along the SWNT axis with a period π/k_F in the spin xy plane (chosen here arbitrarily orthogonal to the SWNT axis).

nanotube (higher values have been reported in [17] though). This compares with the typical energy scales of the electrons, which can be quantified by the value $E_F = v_F k_F/2$ (we set $\hbar = 1$ throughout this Letter), where $k_F/\pi = n_{\rm el}$ is the electron density in the system and v_F ($\approx 8 \times 10^5$ m/s in SWNTs [14,15,24]) is the typical velocity of electron excitations. Through the dependence on $n_{\rm el}$, E_F can vary between the meV to eV range.

Effective model.—Because of the small ratio A/E_F, the energy and time scales related to the electrons and nuclear spins decouple, and we can treat both subsystems separately. A Schrieffer-Wolff transformation of H allows us to obtain an effective Hamiltonian for the nuclear spins [7,8],

$$H_n^{\rm eff} = \frac{1}{2}\sum_{ij\alpha} \frac{J_{ij}^\alpha}{N_\perp^2} \hat{I}_i^\alpha \hat{I}_j^\alpha = \frac{1}{L}\sum_{q\alpha} \frac{J_q^\alpha}{N_\perp^2} \hat{I}_{-q}^\alpha \hat{I}_q^\alpha, \tag{1}$$

where $\alpha = x, y, z$, and $J_{ij}^\alpha = A_0^2 \chi_{ij}^{\alpha\alpha} a/2$ is the effective RKKY [25] interaction between nuclear spins. a is the lattice spacing and provides the short distance cutoff of the continuum theory. The sum over $q = n\pi/L$ for integer n runs over the first Brillouin zone. $\chi_{ij}^{\alpha\alpha} = -ia^{-1}\int_0^\infty dt \langle[\hat{S}_i^\alpha(t), \hat{S}_j^\alpha(0)]\rangle e^{-\eta t}$ (for an infinitesimal $\eta > 0$) is the static electron spin susceptibility. We also have defined $\hat{I}_q^\alpha = \sum_i e^{ir_i q}\hat{I}_i^\alpha$ and $J_q^\alpha = \int dr e^{-irq} J^\alpha(r)$.

The effective electron Hamiltonian, on the other hand, includes the effect of the feedback of the nuclear field on the electrons. Since the spins $\tilde{I} = IN_\perp$ are large, we can choose $H_{\rm el}^{\rm eff} = H_{\rm el} + H_{Ov}$, with $H_{Ov} = \sum_i \mathbf{h}_i \cdot \hat{\mathbf{S}}_i$ and $\mathbf{h}_i = A\langle \hat{\mathbf{I}}_i \rangle$ the nuclear Overhauser field.

Interacting electrons as Luttinger liquid.—We use a bosonized Hamiltonian to describe the interacting electron system of the armchair SWNT, which is naturally in the Luttinger liquid state due to the linear electron dispersion [14,15]. The unit cell of a graphite sheet contains two carbon atoms, which results into a two-band description of the bosonized system. Since mixing between the bands is essentially absent [14,15] we shall, however, focus on a single band only in order to avoid a heavy notation. The bosonized single-band Hamiltonian reads [14,15,26]

$$H_{\rm el} = \sum_{\nu=c,s} \int \frac{dr}{2\pi}\left[\frac{v_\nu}{K_\nu}(\nabla\phi_\nu(r))^2 + v_\nu K_\nu (\nabla\theta_\nu(r))^2\right], \tag{2}$$

where $\phi_{c,s}$ are boson fields such that $-\nabla\phi_{c,s}\sqrt{2}/\pi$ express charge and spin density fluctuations, respectively. $\theta_{c,s}$ are such that $\nabla\theta_{c,s}/\pi$ are canonical conjugate to $\phi_{c,s}$. $v_{c,s} = v_F/K_{c,s}$ are charge and spin wave velocities, and $K_{c,s}$ are the dimensionless Luttinger liquid parameters. For SWNTs [14,15], $K_c \approx 0.2$. If the electron spin SU(2) symmetry is maintained, $K_s = 1$, otherwise $K_s \neq 1$.

Without feedback from nuclear magnetic field.—Let us first assume that there is no feedback from the Overhauser field on the electrons and set $\mathbf{h}_i \equiv 0$. The electron system forms a Luttinger liquid, for which the zero temperature spin susceptibility has a singularity at momentum $q = \pm 2k_F$ induced by backscattering processes [26,27]. At $T > 0$ this singularity turns into a steep but finite minimum: The backscattering part of the spin operator $\hat{S}_i^x$ is expressed in the bosonization language by the operators [26] $\hat{O}_{\rm SDW}^x(r_i) \propto e^{-2ik_F r_i} e^{i\sqrt{2}\phi_c}\cos(\sqrt{2}\theta_s)$, such that $\hat{S}^x = [\hat{O}_{\rm SDW}^x + \hat{O}_{\rm SDW}^{x\dagger}]/2$ plus forward scattering terms. Similar expressions [26] hold for $\hat{S}^y$ and $\hat{S}^z$. We further assume that $J_q^\alpha \equiv J_q$ is isotropic and in particular $K_s = 1$. The correlators between those operators can be evaluated in the standard way and we obtain (for $q > 0$)

$$J_q(g, v_F) \approx -C(g, v_F)(k_B T)^{2g-2}|\Gamma(\kappa)/\Gamma(\kappa + 1 - g)|^2, \tag{3}$$

where $g = (K_c + K_s^{-1})/2$, $\kappa = g/2 - i\lambda_T(q - 2k_F)/4\pi$, depending on the thermal length $\lambda_T = v_F/k_B T$ with k_B the Boltzmann constant. Γ is Euler's Gamma function and $C(g, v_F) = A_0^2 a \sin(\pi g)\Gamma^2(1-g)(2\pi a/v_F)^{2g-2}/8\pi^2 v_F$. We have made the inessential assumption $v_c = v_s = v_F$. Note that J_q is independent of k_F for a linear dispersion. A density dependence of J_q requires a curvature of the electron dispersion, which partially restores Fermi liquid properties [28], a scenario which we disregard for metallic SWNTs. A sketch of J_q is shown in Fig. 2.

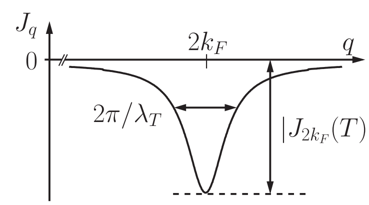

FIG. 2. Sketch of the RKKY interaction J_q given by Eq. (3).

At temperatures $T < T_0^*$ [defined in Eq. (6) below], $|J_{2k_F}(T)| > k_B T$ and the nuclear spins can—classically—minimize the RKKY energy by aligning in a spiral order $\mathbf{I}_i' = IN_\perp[\cos(2k_F r_i)\mathbf{e}_x + \sin(2k_F r_i)\mathbf{e}_y]$, where $\mathbf{e}_{x,y}$ are vectors defining the spin (x, y) plane. We shall henceforth *assume* that this order is established, and show that this assumption is self-consistent. Fluctuations reduce this maximal polarization, and in general $|\langle \hat{\mathbf{I}}_i \rangle| < IN_\perp$. The lowest lying excitations (to order $1/IN_\perp$) in the nuclear spin system are magnons. Since J_{ij} is long ranged the energy cost of local defects, like kinks, scales with the system size and is very high.

For a helimagnet, there exists a gapless magnon band with the dispersion [8] $\omega_q = 2(IN_\perp)(J_{2k_F+q}/N_\perp^2 - J_{2k_F}/N_\perp^2)$. Let $m_i = \langle \hat{\mathbf{I}}_i \rangle \cdot \mathbf{I}_i'/(IN_\perp)^2$ measure the component of the average magnetization along $\mathbf{I}_i'$, normalized to $0 \le m_i \le 1$. Its Fourier component m_{2k_F} acts as an order parameter for the spiral phase. Magnons decrease this order parameter and we have [8]

$$m_{2k_F}(T) = 1 - \frac{a}{(IN_\perp)L}\sum_{q\neq 0}\frac{1}{e^{\omega_q/k_B T} - 1}, \qquad (4)$$

where the sum represents the magnon occupation number. In the continuum limit $L \to \infty$ the integrand is divergent as $1/q^2$ for $q \to 0$ (the $q = 0$ mode is absent because the system is not a ring), showing the absence of true long range order in the 1D system. Despite its appearance the divergence is not a consequence of the Mermin-Wagner theorem [29,30], which forbids long range order in low-dimensional systems for sufficiently short ranged interactions. Since J_{ij} is long ranged this theorem does not apply.

The present situation, however, is very different in that the system has a finite length $L \sim 2\ \mu$m imposed either through the natural length of the nanotube or through an external confining potential. At temperatures $T < T_0^*$ we find that $L \ll \lambda_T$, and so the cost of exciting the first magnon is already very high $\omega_{q=\pi/L} \approx 2I|J_{2k_F}(T)|/N_\perp$. We can define a temperature T_{M0} providing the scale of the excitation of the first magnons by imposing $\omega_q/k_B T \approx 2I|J_{2k_F}(T)|/N_\perp k_B T = 1$. For $T > T_{M0}$ we can then simplify Eq. (4) to

$$m_{2k_F}(T) \approx 1 - \frac{1/IN_\perp}{e^{(T_{M0}/T)^{3-2g}} - 1} \approx 1 - \left(\frac{T}{T_0^*}\right)^{3-2g}, \qquad (5)$$

where we have defined

$$k_B T_0^* = [2I^2 C(g, v_F)\Gamma^2(g/2)\Gamma^{-2}(1 - g/2)]^{1/(3-2g)}. \qquad (6)$$

For the SWNT this temperature satisfies the self-consistency condition $k_B T_{M0} < k_B T_0^* \ll v_F/L$. We use T_0^* as an estimate for the critical temperature. For a typical SWNT T_0^* is very low. With the values given with Fig. 3 we obtain $T_0^* \sim 10\ \mu$K, too low for experimental detection. Yet this analysis completely neglects the feedback of the magnetic field on the electron gas. This leads to a strong renormalization of T_0^*.

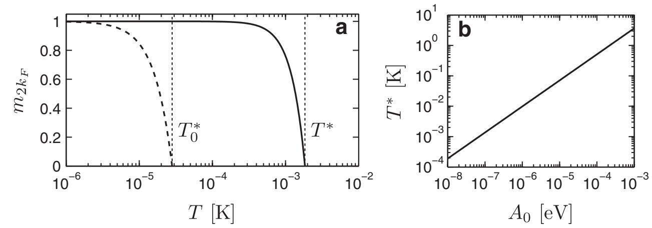

FIG. 3. (a) Magnetization $m_{2k_F}(T)$ [Eq. (5)]. Dashed line: without feedback. Solid line: with feedback. Parameters for the curves are [14,15,23,24] $E_F = 0.1$ eV, $A_0 = 10^{-7}$ eV, $v_F = 8 \times 10^5$ m/s, $a = 2.46$ Å, $K_s = 1$, $K_c = 0.2$ (leading to $g = 0.6$, $g' = 0.33$), and $L = 2\ \mu$m. The vertical lines mark the temperatures written next to them. (b) Characteristic temperature T^* [Eq. (9)] as a function of the hyperfine constant A_0. The curve follows a power law $T^* \propto A_0^{2/(3-2g')} = A_0^{0.86}$, and is plotted up to the self-consistency limit $T^* \approx v_F/Lk_B = 3$ K.

Feedback of nuclear magnetic field on electrons.—The ordering of the nuclear spins leads to a spatially oscillating Overhauser field $\mathbf{h}_i = A\langle \mathbf{I}_i \rangle$ that acts back on the electrons. We choose the electron spin axis such that $\hat{\mathbf{S}} \cdot \mathbf{e}_x = \hat{S}^x$ and $\hat{\mathbf{S}} \cdot \mathbf{e}_y = \hat{S}^y$. The spatial oscillations of $\mathbf{h}_i \propto e^{\pm 2ik_F r_i}$ in H_{Ov} perfectly cancel some of the spatial oscillations of the $\hat{O}_{\mathrm{SDW}}^{x,y}$ operators of the $\hat{S}_i^{x,y}$. Neglecting the remaining (irrelevant) oscillating terms we obtain $H_{Ov} \approx \sum_i A_0 I m_{2k_F} \cos(\sqrt{2K}\phi_+(r_i))$, where we have introduced [31] $\phi_+ = (\phi_c + \theta_s)/\sqrt{K}$ with the normalization $K = K_c + 1/K_s$. The Hamiltonian becomes of the sine-Gordon type and H_{Ov} is relevant in the sense of the renormalization group (RG): The ϕ_+ field is pinned at a minimum of the cosine term of H_{Ov}. The result is a density wave that combines charge and spin degrees of freedom. Fluctuations about the minimum are massive, with a mass associated to an energy scale Δ. At commensurate electron filling umklapp processes would become relevant too, and lead to fully gapped charge and spin sectors. For SWNTs, however, this would require high electron densities leading to $E_F \approx 1.4$ eV. This case is not considered here.

Within a perturbative RG approach we find that

$$\Delta \sim (A_0 I m_{2k_F}/E_F)^{1/(2-g)} v_F/a. \qquad (7)$$

This mass gap Δ is the first important consequence of the feedback. The second important consequence is the spontaneous generation of anisotropy because the spin (x, y) plane is singled out by the Overhauser field. This is seen, for instance, in the spin susceptibilities $\chi^{\alpha\alpha}$. Those can be calculated in the same way as before (details are provided in [31]) if we notice that the massive ϕ_+ field does not contribute to the long-wavelength asymptotics. The finite temperature expressions for the $\chi^{\alpha\alpha}$ are otherwise identical to the case without feedback, and the RKKY couplings J_q^α can be obtained from Eq. (3) upon the following modifications: For χ^{xx} and χ^{yy} the exponent g is replaced by

$g' = 2K_c/K_sK$ and the amplitude is reduced by a factor 2 because a term depending on ϕ_+ only drops out. For χ^{zz} the exponent becomes $g'' = (K_c/K_s + K_cK_s)/2K$ while the amplitude remains unchanged. v_F is replaced by $v_- = (v_c/K_s + v_sK_c)/K$. This leads to

$$J_q^{x,y} = J_q(g', v_-)/2, \qquad J_q^z = J_q(g'', v_-). \tag{8}$$

For $K_c = 0.2$ and $K_s = 1$ we have to compare $g = 0.6$ with the strongly renormalized $g' = 0.33$ and $g'' = 0.17$.

Let us finally note that correlators between ϕ_+, θ_+ can only be neglected as long as $k_BT < \Delta$, i.e., $\lambda_T^{-1} < \xi^{-1}$ with $\xi = v_F/\Delta$ the correlation length. In Eq. (9) below we define a critical temperature T^* similarly to T_0^* before. For $T \ll T^*$, $m_{2k_F} \approx 1$ (see Fig. 3), and we find that $\Delta \gg k_BT$. At $T \to T^*$, however, m_{2k_F} vanishes and so does Δ. The order in electron and nuclear systems, therefore, vanishes simultaneously.

Consequences for magnetization and transport.—The helical order still minimizes the energy and there remains a gapless magnon band [8], $\omega_q = 2I(J'_{2k_F+q} - J'_{2k_F})/N_\perp$, where $J'_q = J_q^x = J_q^y$. The previous discussion of the magnetization remains otherwise unchanged. Replacing J_q by J'_q in Eq. (6) leads to the renormalized critical temperature T^*,

$$k_BT^* = [I^2C(g', v_-)\Gamma^2(g'/2)\Gamma^{-2}(1-g'/2)]^{1/(3-2g')}. \tag{9}$$

The notable difference is the modified exponent. For the parameters displayed with Fig. 3, we obtain the change from $1/(3-2g) = 0.625$ to $1/(3-2g') \approx 0.43$. Quite remarkably this considerably boosts the value of the characteristic temperature from $T_0^* \sim 10\ \mu$K to $T^* \sim 1$ mK. Note that $T^* \ll v_F/Lk_B$ is still satisfied. Figure 3 [(a), solid line] shows the result of the feedback. In Fig. 3(b) we also show the dependence of T^* on A_0, $T^* \propto A_0^{2/(3-2g')} = A_0^{0.86}$.

The order furthermore modifies the transport properties of the system. With the opening of the mass gap in the ϕ_+ channel, half of the conducting modes are blocked and the conductance decreases by the universal factor of 2. As an illustration we consider a SWNT connected to metallic leads. The conductance is given by [32–34] $G = 4e^2/h$, where e is the electron charge, h the Planck constant, and where 4 is the number of conducting channels (2 spin projections and 2 bands). The pinning of the ϕ_+ field (in each band) blocks 2 conductance channels and so reduces the conductance precisely by the factor 2 (see [31] for details). Such a reduction is a direct consequence of the nuclear spin ordering and the Luttinger liquid physics of the electrons, and should be detectable experimentally in standard transport setups.

As a conclusion, we emphasize that the physics described here is quite general and is also relevant for other 1D systems of the Kondo-lattice type.

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