Universal Spin-Flip Transition in Itinerant Antiferromagnets

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We reveal a universal spin-flip (SF) transition as a function of temperature in spin-density-wave (SDW) systems. At low temperatures the antiferromagnetic (AFM) polarization is parallel to the applied field and above a critical temperature the AFM polarization flips perpendicular to the field. This transition occurs in any SDW system and may be considered as a qualitative probe of the itinerant character of AFM in a given material. Our SF transition may provide an explanation to the long-standing puzzle of the SF transition observed in chromium and may be at the origin of the equally puzzling SDW-I to SDW-II transition in Bechgaard salts for which we make experimental predictions.

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The study of itinerant antiferromagnetism (AFM) started in the early 1950s when this state was first observed in chromium [1] and since then it has been a field of continuous interest related to some of the most fascinating problems in materials physics. The first consistent theoretical scheme for itinerant AFM has been elaborated by Overhauser [2] who introduced the spin density wave (SDW) picture. The itinerant character of AFM and the relevance of the SDW picture in chromium are firmly established experimentally [3,4]. Several decades of intense theoretical work led to the construction of a successful microscopic SDW model for chromium [2,5–12]. However, there is still a surprising aspect of the AFM behavior in this material which escapes any microscopic understanding so far. It is the famous spin-flip (SF) transition as a function of temperature for which there are only phenomenological accounts within a Landau framework [4,13–15]. Spin-orbit coupling and dipole-dipole interactions have been shown to be unable to produce a spin-flip transition with temperature [16]. Lacking any microscopic understanding of this first order SF transition, it is viewed up to now as a mysterious peculiarity of chromium.

Other very extensively studied SDW materials are the so-called Bechgaard salts which attracted much interest not only for their SDW behavior, but also for its interplay with superconductivity and related exotic phenomena such as field-induced SDW transitions and quantum-Hall-effect phenomena [17]. These salts are quasi-one-dimensional organic compounds having the form $(TMTSF)_2 - X$ where X denotes a monovalent ion and TMTSF is for tetramethyltetraselenafulvalene [18]. It has been established recently that inside the SDW phase there is a surprising transition to a new SDW phase [19,20]. This SDW-I to SDW-II transition manifests by a sudden change in the T behavior of the NMR relaxation rate from linear just below T_{Neel} to an exponential Arhenius behavior at lower temperatures [19,20]. So far, this phenomenon has been regarded as a peculiar transition from an incomplete SDW state below T_N to a complete SDW state at low T, but such a picture is not consistent with the transport behavior. A spin glass transition has also been invoked [21].

In the present Letter we point out that the spin-flip transition observed in chromium may in fact be a totally generic phenomenon in itinerant AFM. We show that the zero temperature field-induced spin-flop transition in itinerant AFM, at finite temperatures it occurs at a lower critical field and at a sufficiently high temperature it occurs at an arbitrarily small field giving rise to the T-induced spin-flip transition. This SF transition is absolutely generic characterizing any SDW state and therefore should manifest in all itinerant antiferromagnets when crystal fields are negligible. Such a generic behavior of the SDW state has not been noticed so far, probably because in most theoretical works on SDW a one-dimensional framework was adopted lacking the extra spatial dimensions involved in the SF transition. Several aspects of the well-studied SF transition in chromium [4] are in agreement with our SF transition. As for the SDW-I to SDW-II transition in TMTSF's, we predict the identification of a similar SF transition as the one observed in chromium when the measurements of Ref. [22] will be extended to lower temperatures. In fact we argue that the available NMR data [19,20] are totally compatible with our SF transition. Our SF transition can be regarded as a qualitative probe of the itinerant SDW character of AFM in a given material.

We consider the most general mean-field Hamiltonian describing a SDW state in the presence of a uniform magnetic field:

$$H = \sum_{\mathbf{k},\alpha} \xi_{\mathbf{k}\alpha} c^{\dagger}_{\mathbf{k}\alpha} c_{\mathbf{k}\alpha} + \mu_B \sum_{\mathbf{k},\alpha,\beta} c^{\dagger}_{\mathbf{k}\alpha} (\boldsymbol{\sigma} \cdot \mathbf{H}) c_{\mathbf{k}\beta} - \sum_{\mathbf{k},\alpha,\beta} (\boldsymbol{\sigma}\cdot\mathbf{n})_{\alpha\beta} \mathbf{M}_{\mathbf{k}} (\mathbf{c}^{\dagger}_{\mathbf{k}\alpha} \mathbf{c}_{\mathbf{k}+\mathbf{Q}\beta} + \mathrm{H.c.}), \quad (1)$$

where α, β index the spin, $M_{\mathbf{k}}$ is the SDW order parameter that may be anisotropic in some cases, $\mathbf{n}$ defines the axis of the magnetic polarization of the SDW, and $\mathbf{H}$ the Pauli contribution of the applied magnetic field. Orbital effects of the magnetic field are irrelevant in the SDW state. The

electronic dispersion $\xi_{\mathbf{k}}$ considered in the numerical calculations reported here is a tight-binding scheme for a square two dimensional lattice with nearest-neighbors hoping $\xi_{\mathbf{k}} = t(\cos k_x a + \cos k_y a)$. However, our results are independent of the choice of the dispersion as we have verified numerically and discuss later.

To allow for any relative orientation between the SDW polarization and the direction of the field we will use an eight-component spinor formalism. This eight-component space is overcomplete for the present problem, however, it allows us to consider elsewhere the same phenomena in the presence of additional order parameters [23] avoiding a problem dependent formalism. Our space is defined by the eight-component spinor

$$\Psi_{\mathbf{k}}^{\dagger} = (c_{\mathbf{k}\uparrow}^{\dagger} c_{\mathbf{k}\downarrow}^{\dagger} c_{-\mathbf{k}\uparrow} c_{-\mathbf{k}\downarrow} c_{\mathbf{k}+\mathbf{Q}\uparrow}^{\dagger} c_{\mathbf{k}+\mathbf{Q}\downarrow}^{\dagger} c_{-\mathbf{k}-\mathbf{Q}\uparrow} c_{-\mathbf{k}-\mathbf{Q}\downarrow}). \quad (2)$$

The following tensor products provide a convenient basis for the projection of the Hamiltonian in this spinor space

$$\hat{\tau}_i = \hat{\sigma}_i \otimes (\hat{I} \otimes \hat{I}), \qquad \hat{\rho}_i = \hat{I} \otimes (\hat{\sigma}_i \otimes \hat{I}), \qquad \hat{\sigma}_i = \hat{I} \otimes (\hat{I} \otimes \hat{\sigma}_i), \quad (3)$$

where $\hat{\sigma}_i$ are Pauli matrices in usual notations and I the 2×2 identity matrix. This type of multicomponent formalism has been used for the study of magnetic superconductors [24] and recently for the study of excitonic ferromagnetism and colossal magnetoresistance [23].

When $\mathbf{H} \parallel \mathbf{n}$ our Hamiltonian (1) can be written in the eight-component spinor space as follows:

$$\hat{H}_{\parallel} = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^{\dagger} (\xi_{\mathbf{k}} \hat{\tau}_3 \hat{\rho}_3 - M_{\mathbf{k}\parallel} \hat{\tau}_1 \hat{\rho}_3 \hat{\sigma}_3 + \mu_B H \hat{\rho}_3 \hat{\sigma}_3) \Psi_{\mathbf{k}}. \quad (4)$$

The Green's function corresponding to this Hamiltonian is now an 8×8 matrix which can be shown to take the following form in our representation:

$$\hat{G}_{\parallel}(\mathbf{k}, i\omega_n) = -[i\omega_n + \xi_{\mathbf{k}} \hat{\tau}_3 \hat{\rho}_3 - M_{\mathbf{k}\parallel} \hat{\tau}_1 \hat{\rho}_3 \hat{\sigma}_3 + \mu_B H \hat{\rho}_3 \hat{\sigma}_3][\omega_n^2 + \xi_{\mathbf{k}}^2 + M_{\mathbf{k}\parallel}^2 + \mu_B^2 H^2 - 2\xi_{\mathbf{k}} \mu_B H \hat{\tau}_3 \hat{\sigma}_3 + 2M_{\mathbf{k}\parallel} \mu_B H \hat{\tau}_1] \times [\omega_n^2 + E_{+\parallel}^2(\mathbf{k})]^{-1} [\omega_n^2 + E_{-\parallel}^2(\mathbf{k})]^{-1}, \quad (5)$$

where

$$E_{\pm\parallel}(\mathbf{k}) = \sqrt{\xi_{\mathbf{k}}^2 + M_{\mathbf{k}\parallel}^2} \pm \mu_B H. \quad (6)$$

The SDW gap equation results from the self-consistency relation $M_{\mathbf{k}\parallel} = \frac{1}{8} T \sum_{\mathbf{k}'} \sum_n V_{\mathbf{k}\mathbf{k}'} \times \mathrm{Tr}\{\hat{\tau}_1 \hat{\rho}_3 \hat{\sigma}_3 \hat{G}_{\mathbf{k}'n\parallel}\}$ and after analytic summation over the Matsubara frequencies it can be shown to take the following form:

$$M_{\mathbf{k}\parallel} = \sum_{\mathbf{k}'} V_{\mathbf{k}\mathbf{k}'} \frac{M_{\mathbf{k}'\parallel}}{4\sqrt{\xi_{\mathbf{k}'}^2 + M_{\mathbf{k}'\parallel}^2}} \times \left[\tanh\left(\frac{E_{+\parallel}(\mathbf{k}')}{2T}\right) + \tanh\left(\frac{E_{-\parallel}(\mathbf{k}')}{2T}\right) \right], \quad (7)$$

which is identical with the gap equation of a singlet BCS superconductor in a Zeeman field. The field $\mu_B H$ appears only in the hyperbolic tangent functions and in the zero temperature regime we have $|\tanh(E_{\pm\parallel}(\mathbf{k})/2T)| \approx 1$. Therefore a magnetic field smaller than the critical field and parallel to the polarization of the SDW has practically no influence on the SDW in the zero temperature regime. On the other hand, when the field is sufficiently large $\mu_B H > M_{\mathbf{k}\parallel}$ then in the $T \to 0$ regime $\tanh(E_{-\parallel}(\mathbf{k})/2T) = -\tanh(E_{+\parallel}(\mathbf{k})/2T) = -1$ and the SDW is eliminated. Therefore, in the $T \to 0$ regime there is a critical magnetic field parallel to the polarization of the SDW ($\mu_B H_c \approx M_{\mathbf{k}\parallel}$ if the gap is isotropic) that can melt the SDW. This is the analog of the well known Clogston-Chandrasekhar critical field [25] in superconductivity which has indeed been observed in superconducting films when the field is applied parallel to the film planes [26].

Numerical solutions of the gap equation confirm this behavior [see Fig. 1(a)]. Indeed, in the zero temperature regime $M_{\mathbf{k}\parallel}$ as a function of the field has a steplike behavior and for $\mu_B H > M_{\mathbf{k}\parallel}$ [in the example shown in Fig. 1(a) the gap is isotropic], $M_{\mathbf{k}\parallel}$ is identically zero. The melting of the SDW when $\mu_B H_c = M_{\mathbf{k}\parallel}$ manifests already in the structure of the poles of the Green's function reported in (6). One of the two quasiparticles poles $E_{-\parallel}(\mathbf{k})$ moves to zero when $\mu_B H_c = M_{\mathbf{k}\parallel}$ and there is no gap on the Fermi surface. Because in $E_{-\parallel}(\mathbf{k})$ the field $\mu_B H$ and the SDW gap $M_{\mathbf{k}}$ contribute into terms which have opposite sign we can say that the $\mathbf{H} \parallel \mathbf{n}$ magnetic field is in direct competition with the SDW. The situation will be shown below to be different if the polarization of the SDW is perpendicular to the field.

We now consider the case in which $\mathbf{H} \perp \mathbf{n}$. In the same eight-component formalism our Hamiltonian (1) can be written as follows:

$$\hat{H}_{\perp} = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^{\dagger} (\xi_{\mathbf{k}} \hat{\tau}_3 \hat{\rho}_3 - M_{\mathbf{k}\perp} \hat{\tau}_1 \hat{\rho}_3 \hat{\sigma}_3 + \mu_B H \hat{\rho}_3 \hat{\sigma}_1) \Psi_{\mathbf{k}}. \quad (8)$$

The corresponding matrix Green's function diagonalized in our representation takes the following form:

$$\hat{G}_{\perp}(\mathbf{k}, i\omega_n) = -[i\omega_n + \xi_{\mathbf{k}} \hat{\tau}_3 \hat{\rho}_3 - M_{\mathbf{k}\perp} \hat{\tau}_1 \hat{\rho}_3 \hat{\sigma}_3 + \mu_B H \hat{\rho}_3 \hat{\sigma}_1][\omega_n^2 + \xi_{\mathbf{k}}^2 + M_{\mathbf{k}\perp}^2 + \mu_B^2 H^2 - 2\xi_{\mathbf{k}} \mu_B H \hat{\tau}_3 \hat{\sigma}_1] \times [\omega_n^2 + E_{+\perp}^2(\mathbf{k})]^{-1} [\omega_n^2 + E_{-\perp}^2(\mathbf{k})]^{-1}, \quad (9)$$

and the quasiparticles poles are now defined by

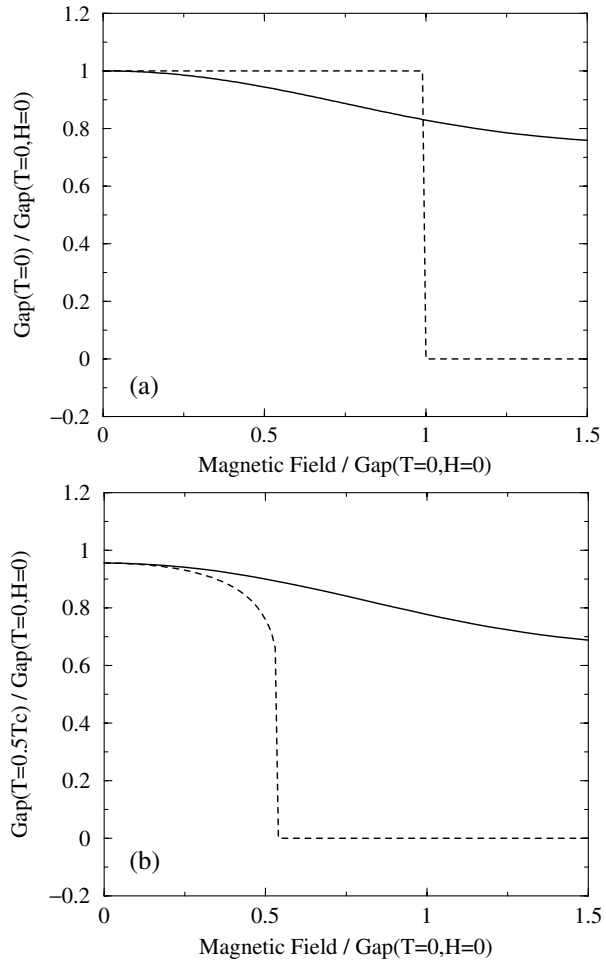

FIG. 1. (a) Evolution of the SDW gap as a function of the magnetic field in the zero temperature regime when the SDW polarization is parallel (dashed line) or perpendicular (full line) to the field direction. At low fields the parallel polarization has a lower free energy (higher SDW gap). When $\mu_B H$ exceeds the gap, only the perpendicular polarization has a finite gap leading to the spin-flop transition from parallel (at low fields) to perpendicular SDW polarization as a function of the field. (b) Same as in (a) but at a finite temperature $T = 0.5T_N$. The spin-flop transition is eliminated.

$$E_{\pm\perp}(\mathbf{k}) = \sqrt{(\xi_{\mathbf{k}} \pm \mu_B H)^2 + M_{\mathbf{k}\perp}^2}. \tag{10}$$

From the structure of the poles it is already obvious that the perpendicular field is not in direct competition with the SDW. None of the quasiparticle poles given in (10) can be set to zero no matter how large the magnetic field is. This indicates that the magnetic field cannot melt the SDW. Let us check this by calculating the gap equation which can now be shown to take the following form:

$$M_{\mathbf{k}\perp} = \sum_{\mathbf{k}'} V_{\mathbf{k}\mathbf{k}'} M_{\mathbf{k}'\perp} \left[\frac{1}{4E_{+\perp}(\mathbf{k}')} \tanh\left(\frac{E_{+\perp}(\mathbf{k}')}{2T}\right) + \frac{1}{4E_{-\perp}(\mathbf{k}')} \tanh\left(\frac{E_{-\perp}(\mathbf{k}')}{2T}\right) \right]. \tag{11}$$

Only in the limit $\mu_B H \to \infty$ the gap equation provides an identically zero solution. Moreover, any finite perpendicular field reduces gradually the SDW gap (because it appears in the denominator) even in the $T \to 0$ regime no matter how small it is in sharp contrast with the parallel field behavior where in the $T \to 0$ regime fields smaller than the gap have practically no influence.

The above behavior in the $T \to 0$ regime is verified by numerical solutions as shown in Fig. 1(a). Therefore, if the polarization $\mathbf{n}$ of the SDW is free as in any perfectly itinerant SDW system, we naturally expect the following behavior of $\mathbf{n}$ in the presence of a field in the $T \to 0$ regime. For weak fields the polarization of the SDW will chose the direction parallel to the field since in that way it is insensitive on it. As the field grows, and as long as it remains smaller than the gap, $\mathbf{n}$ remains locked parallel to the direction of the field. When the field equals the gap, the SDW will suddenly flip its polarization from $\mathbf{n} \| \mathbf{H}$ to $\mathbf{n} \perp \mathbf{H}$. This first order transition illustrated in Fig. 1(a) is the itinerant counterpart of the well studied spin-flop transition in the localized magnetism picture. However, the situation is qualitatively different here. In fact, in the localized magnetic moments picture, at any finite field the moments have a tendency to be perpendicular to the field, while here this tendency exists only above the critical field.

More suprising, and without counterpart in the localized limit, is the behavior of our spin-flop transition at finite temperatures. Finite temperature solutions of the gap equations indicate that although at low temperatures and low fields the $\mathbf{n} \| \mathbf{H}$ state prevails [see Fig. 1(a)], at a higher temperature $T \approx T_N/2$ the $\mathbf{n} \perp \mathbf{H}$ state prevails whatever the field [Fig. 1(b)]. Above a given temperature, the magnetic field-induced spin-flop transition is in fact eliminated. In fact, the condensation free energy is a growing function of the gap and consequently the states with the higher gap have also the lowest free energy. An example of the evolution of both $M_\parallel$ and $M_\perp$ as a function of the field and the temperature is reported in Fig. 2. The zero temperature spin-flop transition from $\mathbf{n} \| \mathbf{H}$ to $\mathbf{n} \perp \mathbf{H}$, by rising the temperature it appears at lower critical fields and at $T \approx 0.435T_N$ the critical field of this transition is zero. The physical origin of this thermally induced spin-flip (SF) transition is probably related with the phase space for spin fluctuations. In fact, with the SDW polarization perpendicular to the field, the available phase space for thermal excitation of the spins is larger than in the case of a polarization parallel to that of the field. At a sufficiently high temperature this phase-space gain apparently dominates inducing the SF transition.

Our SF transition displays as a function of the field and the temperature many of the characteristics of the SF transition in chromium [4] which we believe is its most obvious physical realization. We must note here that in chromium the spin-flip transition is usually discussed with respect to the direction of the CDW wave vector $\mathbf{Q}$ and not of the magnetic field. However, a sharp first order

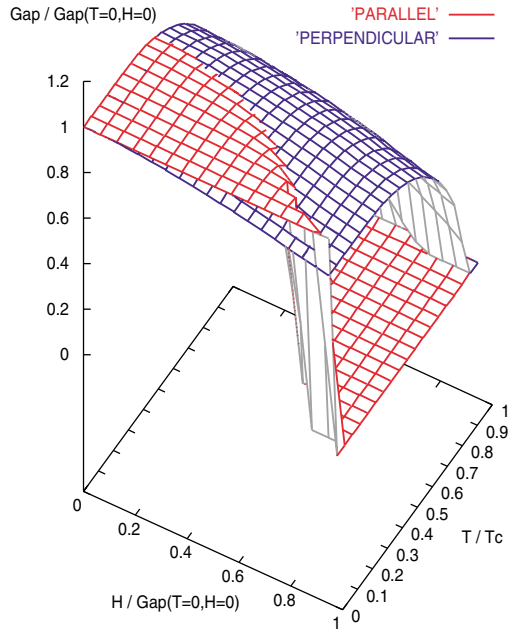

FIG. 2 (color online). Evolution of the SDW gap as a function of the magnetic field and temperature for parallel (red) and perpendicular (blue) to the field polarization of the SDW. At low fields and temperatures the parallel polarization prevails.

transition is observed only when the field and **Q** are parallel, and as our analysis points out, it is in fact the direction of the field that matters. The relevance of our picture is further supported by the fact that the order of magnitude of the ratio T_{SF}/T_N in chromium is just in the range in which we predict this transition should happen. Moreover, when particle-hole asymmetry is introduced including, for example, a next nearest-neighbors hopping term in our dispersion and reducing thus the nesting, our T_{SF}/T_N is reduced and this precisely what is observed by alloying chromium [4]. The most likely range of this transition is $0.20T_N \leq T_{SF} \leq 0.45T_N$, the highest value being indicative of a particle-hole symmetric system. In bare chromium $T_{SF} \approx 0.395T_N$ which is just in the range where we expect our SF transition.

As for the SDW-I to SDW-II transition in Bechgaard salts, here as well it happens precisely in the temperature range in which we predict our SF transition ($\approx T_{\mathrm{Neel}}/3$). Moreover, the Arhenius low-T behavior of the NMR relaxation rate [19,20] is consistent with **n** locked parallel to the field while the linear Korringa behavior at higher T and up to T_{Neel} is consistent with both **n** perpendicular to the field and the observed insulating transport behavior. We predict that extending the measurements of [22] to temperatures below 4 K could definitely establish the SF character of the SDW-I to SDW-II transition which is observed at about 3.5 K in Bechgaard salts.

In conclusion, we have shown that in all SDW systems occurs a SF transition by varying temperature. In the low-Tphase the SDW polarization is parallel to the field while above the SF transition it is perpendicular. This SF transition has been identified in chromium and is likely to be the origin of the SDW-I to SDW-II transition in Bechgaard salts. It represents the fingerprint of the itinerant character of AFM in a given material.

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