J. Phys. Soc. Jpn. **52** (1983) Suppl. p. 117–120

ON THE REMANENT MAGNETIZATION IN CuMn

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Recent experiments have revealed that the remanent magnetization in CuMn (and in similar alloys) involves a peculiar aspect of instability that seems beyond proposals to account for from the current spin-glass theories. After a summary of the reported facts available, we present a model based on a conduction-electron spin instability. (A full account of the present studies is to be published [13].)

1. Experimentally Reported Facts

Static magnetizations - Old results of Schmitt-Jacobs [1] and Kouvel [2]

The concepts of isothermal remanent (IRM) and thermo-renament (TRM) magnetization were present in the early experiments performed by these authors before the spin-glass phase has been recognized. Moreover, a remarkable asymmetry of the hysteresis M-H cycle of TRM, when the system is cooled in presence of a magnetic field, was described by Kouvel. (In general, TRM $\geq$ IRM and the equality holds when IRM is prepared by an infinitely strong field, called the saturation (S)TRM. The asymmetry is such that the center of the cycle is shifted to the opposite direction to the cooling field of TRM.) Another point of interest manifest in these experiments is, though not discussed there or even left unnoticed afterwards, is a scaling characteristic:

(1) The dependence of STRM σ_r on the Mn concentration is that the low C regime ($C <\sim 2\%$Mn) $\sigma_r \propto C$, and numerically

$$\sigma_r \simeq 0.3 \text{ emu/cc, C\% } (0.04\mu_B/\text{Mn by S} = 2) \qquad (1.1)$$

the high C regime ($2\% < C < 10\%$) $\sigma_r \propto C^{2-\varepsilon}$ (The spin-glass characteristic is lost above $C \sim 10\%$ [2].)

New aspects reported by Monod, Préjean, Tissier [3] and Préjean, Joliclerc, Monod [4]. Main features are as follows:

(2) The hysteresis cycles in the low C regime involve a very sharp reversal of σ_r and are square shaped, implying a short duration (less than second) of the reversal and its near completeness. It has been described adopting the Landau-Lifshitz model of rigid dipole subject to a rotary instability with a varying external field. It necessitates that a domain-theoretic explanation of the phenomena is inadequate, because in such a regime the system is essentially in a single domain.

(3) σ_r, once formed and when the external field H is removed or varied to the opposite direction, becomes unstable (maybe in a metastable state) subject to a logarithmic time decay. The reversal time and the reversal field are sensitive to alloying with third, nonmagnetic impurities depending largely on the impurity atom spin-orbit strength. This suggests the conduction (s-) electron spin-lattice rate to prevail the instability. A related fact is that no such instability exists in the M-H curve for AuFe whose electronic structure is similar to CuMn apart from the largeness of the spin-orbit effect.

(4) Some remarkable temperature characteristics exist: For TRM, the magnitude of σ_r changes with the temperature of the field cooling, but the width ΔH of

the hysteresis cycle also changes so that

$$\sigma_r \Delta H = 4K \text{ (K: "anisotropy energy")} = \text{nearly independent of temperature.} \qquad (1.2)$$

For IRM, temperature variations in the course of a hysteresis cycle do not change the magnitude σ_r but change the displacement field H_d (the center ordinate of the cycle) such that the negative displacement increases by a cooling — this is not a field cool due to the smallness of H_d.

Supplementary information from dynamical experiments The zero field NMR on Cu in CuMn performed by Alloul [5] revealed a resolved spin-echo spectrum due to 1st and 4th neigbor Mn sites, a direct evidence of the very strong (RKKY) exchange field acting on Cu sites ($\sim 10^5$ gauss for 1% Mn in the electronic unit). Including the non-zero field NMR results, Alloul was able to confirm the rigid-dipole instability model, emphasizing the monodomain nature of the magnetism. This led him to perform static transverse susceptibility measurements [6] to examine more details of the model. On the other hand, ESR by Monod and Berthier [7] shed a light on the nature of the "antiferro resonance" which has long been puzzling: it is essentially the ESR of TRM subject to a rigid rotation and instability where the anisotropy field (1.2) also plays a role of determining the resonance frequency. (The zero-field cooled case of ESR is still mysterious.) For more details, see Schultz et al [8] who demonstrated a clear C^2 scaling of K and its temperature dependence.

Thus, it would be an urgent question to construct a model, consistent with all the main features above, to understand the origin of the rigid dipole formed macroscopically in the spin-glass state. Our basic starting point in what follows is that the RKKY coupled d-spin system is not the all to form the remanent magnetization in CuMn but rather that the conduction electron spins in the Fermi shell act primarily.

2. A Retreatment of the s-d interaction

The Kasuya-Yosida theory of the s-d interaction [9][10] contains a variational treatment i.e. free energy minimum of a part of the s-d interaction, diagonal in regards the wave vectors of the s-electrons, which determines the s-magnetization uniform in space. The off-diagonal parts are subsequently taken into account by the usual perturbation approach to second order. Consequently, Kasuya derived three types of the effective spin coupling; an indirect exchange known as RKKY, the pseudo-scalar type and the residual s-d type (a term involving the Kondo type logarithmic singularity [11]); the latter two types of the effective coupling depending still on the s-spin densities and hence active only on the Fermi shell of a width $2\Delta E$ ($\propto M_s$) singly occupied.

Thus, we reformulate the perturbation theory on the s-d system for free energy in such a way that the perturbation is performed first, under a suitable, yet undetermined distribution for both spin directions, and then the resulting free-energy expression is minimized. To be adapted for the spin-glass state, moreover, a relevant renormalization procedure is necessary so that the RKKY interaction is incorporated into the unperturbed part of the hamiltonian. A consistent formulation is possible by dividing the s-d hamiltonian into two parts i.e. the high frequency part and the low frequency part (see Fig.(1)):

$$(S) \quad \begin{cases} \mathcal{H}_{sd}(\text{low}) : \text{with those matrix elements } J_{kk'} \text{ such that} \\ \qquad |\varepsilon_k - \varepsilon_F| \leq \Delta E , \quad |\varepsilon_{k'} - \varepsilon_F| \leq \Delta E ; \qquad (2.1) \\ \mathcal{H}_{sd}(\text{high}) : \text{with } J_{kk'} \text{ of k and k' all other than (2.1).} \end{cases}$$

The consistency is assured by incorporating $\mathcal{H}_{sd}$(high) into the unperturbed part as the RKKY (by a canonical transformation similar to the BCS theory), where the smallness of $\Delta E(\sim 10^{-3}\varepsilon_F)$ retained will not modify the short-range part of the RKKY coupling, and by calculating the 2nd order perturbation of $\mathcal{H}_{sd}$ (low) with each d-spin being assumed as acted on by the strong molecular field.

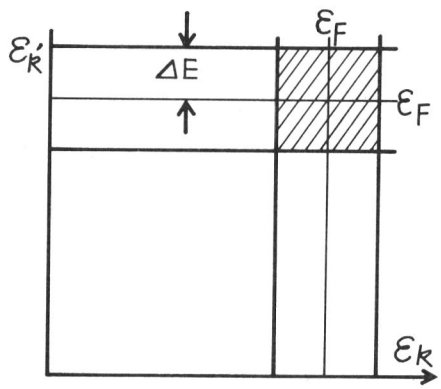

Fig.1. Specification of the matrix elements $J_{kk'}$ to be included into the low-and high-frequency parts $\mathcal{H}_{sd}$. The shaded area is denoted by (S).

A significant effect that arises from including the molecular field as the propagation factor of the individual d spins in conjunction with the kinetic propagation of the Fermi-shell spin density in $\mathcal{H}_{sd}(\text{low})$, characteristic of the spin-glass regime because of the absence of the molecular field for $T > T_g$, is represented by

$$\mathcal{H}_A^{(2)} = -E_A \sum_{k,k'}^{(s)} \frac{J_{kk'}^2}{2N^2} \left[\frac{f_k + f_{k'} - f_k f_{k'} + (f_{k_+} - f_{k_-})(f_{k'_+} - f_{k'_-})}{(g\mu_B H_E)^2 - (\varepsilon_k - \varepsilon_{k'})^2} \right] \tag{2.2}$$

with $$E_A = \sum_n S_n^{z(n)} g\mu_B H_{En}^{z(n)} \quad , \quad f_k = f_{k_+} + f_{k_-} \tag{2.2a}$$

and $$\mathcal{H}_B^{(2)} = -E_B \sum_{k,k'}^{(s)} \frac{J_{kk'}^2}{N^2} \left[\frac{f_{k_+} - f_{k_-}}{(g\mu_B H_E)^2 - (\varepsilon_k - \varepsilon_{k'})^2} \right] \tag{2.3}$$

with $$E_B = \sum_n (S_n^{z(n)})^2 g\mu_B H_{En}^{z(n)} \quad , \tag{2.3a}$$

where H_{En} stands for the molecular field acting on the d-site n. These are the two kinds of the 2nd order exchange self-energy; the terminology due to Orback and Spencer, given to the ordinary uniform Zeeman field [12]. An important difference between their formula and (2.2,3) is that the summation k,k' in the latter is restricted to (S) specified by (2.1), because the molecular field acting as if it were the Zeeman field actually stems from the isotropic RKKY coupling and should not be present to propagate $\mathcal{H}_{sd}(\text{high})$ to derive $\mathcal{H}_{RKKY}$ itself.

Another point of speciality in the above formulas must be mentioned. The molecular field axis depends generally on the individual d-spins. The quantization axis of the Fermi shell spins cannot accordingly be well specified. So, we shall adopt the following simple assumptions that may conform, as the roughest allowance, to the actual spin-glass situation: a field cooled state (TRM formed): Every molecular field axes are parallel to (or, distributed at least centering at) the direction of the cooling field. Then, the Fermi distribution function $f_{k\pm}$ with the specified spin direction makes sense as usual, and in particular at absolute zero temperature

$$f_k = 1 \quad \text{and} \quad f_{k_+} - f_{k_-} = \text{sgn}(\Delta E) \quad , \quad k\varepsilon(S) \tag{2.4}$$

the zero-field cooled state : The molecular field axes are directionally completely uniform, but the situation is different from the paramagnetic state in that the existence of the molecular field in a particular direction means the existence of an ordered phase in the groupwise sense : (2.4) is then still applicable to each group. By summation all over the groups, however,

$$\mathcal{H}_B^{(2)} = 0 \quad \text{or simply} \quad E_B = 0 \tag{2.5}$$

Note that even in this state $E_A \neq 0$, in fact $E_A > 0$ to lower the magnetic free energy. Note also that the no-net-magnetization implies

$$\sum_n H_{En} = 0 \quad \text{(characteristic of the spin-glass state).} \tag{2.6}$$

One more simplifying assumption that $|H_{En}| = H_E$ (already meant in (2.2,3)) enables us to carry out the double summation k,k' to give

$$\mathcal{H}_A^{(2)} + \mathcal{H}_B^{(2)} = -(a + b\,\mathrm{sgn}(M_s))\Phi(M_s) \;;\; \Phi(x) = \frac{1}{2}\begin{pmatrix}(x+p)\log|1+x/p| \\ -(x-p)\log|1-x/p|\end{pmatrix}$$

with $P = \frac{g}{2}\chi_p H_E$ (χ_p : Pauli para susceptibility) (2.7)

and $a = 2(\lambda\chi_p)^2 \frac{1}{p}\sum_n M_n^z H_{En}^z$, $b = 2(\lambda\chi_p)^2 \frac{1}{p}\sum_n M_n^z S_n^z H_{En}^z$. (2.8)

It is now ready to calculate the magnetization and to show its instability at absolute zero temperature where $M_d = 0$ (d-spins are frozen)

$$F(M_s) = \frac{M_s^2}{2\chi_p} - H\cdot M_s - (a + b\,\mathrm{sgn}(M_s))\Phi(M_s) \tag{2.9}$$

$$M_s \longmapsto F'(M_s) = 0 \text{ i.e. } M_s = x = \chi_p(H+(a+b\,\mathrm{sgn}(x))\Phi'(x). \tag{2.10}$$

The nonlinear equation (2.10) for x is shown to have a root $x(\neq 0)$ for $H = 0$ in a branch of $\Phi(x)$ and to exhibit a bistable transition from the branch corresponding to $x(H=0) = 0$ to the above. Further, if F is considered as the function of $\theta = M_s \hat{} H$, it causes a rotary instability of the Landau-Lifshitz type (Fig.2).

3. Two-Component Thermodynamics of s-d Magnetizations

To examine the actual features of the observations, it is necessary to extend the above idea to non-zero low temperatures ($T \ll T_g$): The relevant free energy is, then

$$F(M_s,M_d) = \frac{M_s^2}{2\chi_p} + \frac{1}{2}\chi_d^{-1} M_d M_d - H\cdot(M_s + M_d) - \lambda M_s\cdot M_d - K(M_{s//}) \tag{3.1}$$

where $K(M_{s//})$ represents the 2nd order exchange self-energy (2.7) with a complex order parameter, $X = p + i\Gamma$ ($\Gamma \propto$ spin-lattice rate). Our consideration of the Fermi-shell instability results in

$$\sigma_r = \chi_p|1 + \lambda\chi_{d//}|\, H_E. \tag{3.2}$$

This is shown to fit in the magnitude and scaling characteristic (1). Detailed aspect how (1)∿(4) can be understood is left to our publication [13].

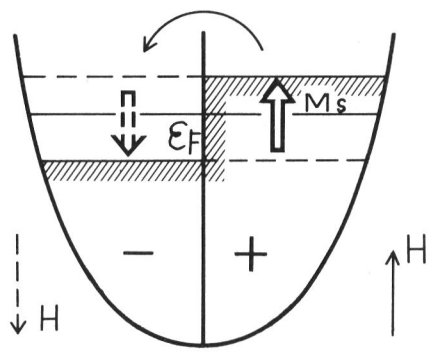

Fig.2. Illustration of the instability of M_s reverse by rotation.

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