Possibility of transition of paramagnetic crystals into the "spin glass phase"

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It is shown that in adiabatic demagnetization in a rotating coordinate frame (ADRCF) in magnetically diluted paramagnetic crystals, a transition to the "spin-glass" phase can be observed. The conditions of the transition are obtained and a scheme is proposed for the appropriate experiment.

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One of the most interesting results of recent years in the region of magnetism is the attainment of dipole ferromagnetic and antiferromagnetic ordering in nuclear spin systems with the aid of ADRCF.[1] Inasmuch as the concept of spin temperature in a rotating coordinate frame (RCF) under certain conditions is valid also for magnetically diluted electronic spin systems,[2] the question arises of the possibility of observing similar effects in objects that differ principally in the random spatial distribution of the spins over the sample.

According to Weiss's simplest model of the molecular field, the temperature of a ferromagnetic transition due to dipole interactions is $T_c = 4\pi\mu^2 N/3k \sim \hbar\delta/k$

where μ is the spin magnetic moment, N is the number of spins S per unit volume, and δ is the high temperature dipole width of the paramagnetic-resonance line. On the other hand, the minimum absolute value of the RCF spin temperature, which is attained at the instant when the saturating field passes through the center of the resonance curve, is[3]

$$T_{min} \approx \frac{T_0\sqrt{M_2}}{\gamma H_0\sqrt{3}}, \tag{1}$$

where T_0 is the lattice temperature, H_0 is the external magnetic field, γ is the gyromagnetic ratio, and M_2 is the second moment of the paramagnetic resonance. Comparing T_c with T_{min} and recognizing that in magnetically diluted systems $\sqrt{M_2}/\delta \sim 1/\sqrt{c} \gg 1$,[3] we can easily verify that in order to attain T_c at typical paramagnetic-impurity concentrations $c \sim 10^{-2}$—10^{-4} it is necessary to have practically unreachable values of H_0/T_0 (thus at $S=1/2$, $c=10^{-3}$, and $T_0 = 1\,°K$ we need $H_0 \geq 4\times 10^5$ Oe).

The situation changes radically if account is taken of the possibility of a fundamentally new type of magnetic ordering, the so-called "spin glass,"[4] in which the spins are "frozen" in local fields, but owing to their random orientation they produce no macroscopic spontaneous magnetization (a phase transition into this state was recently observed in metal alloys of the AuFe type[5]). As shown in[6], the condition for the existence of the spin-glass phase in such alloys is

$$\bar{J}_{ij} \ll (\overline{J_{ij}^2})^{1/2}, \tag{2}$$

where J_{ij} is an exchange integral determined by the long-range interaction via the conduction electrons (the "RKKY mechanism"), and the superior bar denotes averaging over the volume. Owing to the rapidly oscillating sign-alternating dependence of J_{ij} on the distance r_{ij} between spins (which is inherent in the RKKY potential), in conjunction with the random distribution of the "magnetic" atoms, we have $\bar{J}_{ij} \approx 0$ and this ensures satisfaction of (2).

We now turn to impurity paramagnetic crystals and write down the dipole-dipole interaction Hamiltonian in the RCF[3]:

$$\mathcal{H}_d^{RCF} = \sum_{i<j} A_{ij}\,(2S_i^z S_j^z - S_i^x S_j^x - S_i^y S_j^y),$$

where the z axis is directed along $\mathbf{H}_0$ and x along the rotating high-frequency field $\mathbf{H}_1$, with

$$A_{ij} = \frac{\gamma^2\hbar}{2r_{ij}^3}(1 - 3\cos^2\theta_{ij}), \tag{3}$$

where θ_{ij} is the angle between $\mathbf{H}_0$ and $\mathbf{r}_{ij}$. It is seen from (3) that A_{ij}, unlike the RKKY potential, is not an oscillating function of $\mathbf{r}_{ij}$, but is instead a sign-alternating function of θ_{ij}, and averaging over the random angles again gives $\bar{A}_{ij} \approx 0$ (for cubic lattices this is an exact equality). Thus, there is an essential analogy between alloys and magnetically diluted paramagnetic crystals; it is easy to see in the latter case the condition (2) reduces to the requirement $\delta \ll \sqrt{M_2}$, which is certainly satisfied at sufficiently small c.[3] We note that this inequality, while ensuring the possibility of the spin-glass phase, hinders at the same time, as already noted, the ferromagnetic transition in ADRCF.

To estimate the temperature T_{sg} of the phase transition we use a simplified approach based on the molecular-field model.[7] Noting that the form of $\mathcal{H}_d^{RCF}$

makes it possible to set up independent equations for each spin component,[8] and recognizing that at the instant when T_{min} is reached the static field in the RCF is equal to zero, we write (for $S=1/2$)

$$|\langle S_i^z\rangle| = -\frac{\hbar}{2}\,\mathrm{th}\left\{\frac{1}{kT}\sum_j\left[\mathrm{sign}\left(\frac{\langle S_i^z\rangle}{\langle S_j^z\rangle}\right)\right]A_{ij}\,|\langle S_j^z\rangle|\right\}, \tag{4}$$

where $|\ldots|$ is the modulus and $\langle\ldots\rangle$ the thermodynamic mean value. In the equations for S^x and S^y it is necessary to add on the right-hand side the factor $(-1/2)$.

Proceeding as in[7], i.e., expanding (4) in a series about T_{sg}, iterating, and averaging over the volume under the assumption of no correlation between the the spin orientations, we can easily obtain for the order parameter $\overline{\langle S_i^a\rangle}$ an equation that has a nonzero solution at

$$T \leqslant T_{sg}^z = \frac{\hbar}{2k}\left(\overline{\sum_j A_{ij}^2}\right)^{1/2} = \frac{\hbar}{3k}\sqrt{M_2}\ . \tag{5}$$

For S^x and S^y, the value of T_{sg} is reduced by one half.

Comparing (1) with (5), we obtain the condition for the transition to the spin-glass phase in ADRCF:

$$\frac{\gamma\hbar H_0}{kT_0} \geqslant \sqrt{3}\ .$$

This condition can be relatively easily obtained in experiment—for example, at $T_0 = 1$ °K and $H_0 \geq 13$ kOe; furthermore, and this is particularly interesting, it does not depend theoretically on c (in practice, a concentration dependence should arise, however, owing to the influence of the inhomogeneous EPR broadening and the spin-lattice relaxation).

Following[7], we can calculate the critical behavior of the longitudinal magnetic susceptibility $\chi_{\parallel}(T)$ and obtain the discontinuity of the derivative at $T = T_{sg}$, but the details of the calculation are very sensitive to the choice of the theoretical model: we confine ourselves therefore at this stage to a reference to the experimental results for alloys in which a sharp peak of $\chi_{\parallel}$ was actually observed.[5] When performing the appropriate experiment on paramagnetic crystals it must be taken into account that the "lifetime" of the spin-glass phase in the RCF is limited by the spin-lattice relaxation (10^{-1}—10^{-3} sec), so that the registration of $\chi_{\parallel}$ should be carried out simultaneously with ADRCF procedure ("in flight"). The procedure described in[9] is convenient for such measurements.

Thus, the possibility of observing a transition of paramagnetic crystals into a spin-glass phase in ADRCF is perfectly realistic. Such an experiment will add greatly to the physical picture of the behavior of spin systems at low temperatures.

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