the high-temperature regime where perturbative treatments are adequate.

In conclusion, we have demonstrated that the mapping of an easy-plane ferromagnet in a field to sG is inadequate for the parameters where the neutron-scattering experiments[7] in $CsNiF_3$ have been performed. This is because the out-of-plane spin fluctuations turned out to be of crucial importance, so that a nearly isotropic model [Eq. (3), $A \approx 0$] represents a much better approximation. Accordingly, the observed low-frequency resonance in $S_{xx}(q,\omega)$ (Ref. 7 and Fig. 3) must be attributed to multimagnon difference processes or pulse-soliton features of this model.

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Simultaneous ESR and Magnetization Measurements Characterizing the Spin-Glass State

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Two ESR modes in CuMnNi spin-glass alloys are observed. Both modes are interpreted with a phenomenological theory incorporating an order parameter, remanent magnetization, and anisotropy energy. From simultaneous magnetization and ESR measurements, it has been possible to deduce the temperature and concentration dependence of the anisotropy constant, which has interesting scaling properties. The angular dependence of the field-cooled ESR and magnetization data suggest the need for another order parameter which vanishes at T_g.

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We have applied a simple technique[1] to perform *simultaneous* magnetization and ESR measurements on the CuMn spin-glass system as a function of manganese concentration C_{Mn} (atomic percent), temperature T, spectrometer radian frequency ω, remanent magnetization M_r (via dc field for cooling H_c), θ (orientation of dc field to $\vec{H}_c$), concentration of an additional impurity C_i, and sample preparation and geometry. Most of the data will be reported elsewhere, but several features which present new insights into the spin-glass state will be discussed.

In particular we report the observation of a second ESR mode of the system. We are able to interpret the frequency dependence of this new mode, as well as the one usually observed, in terms of a model free energy incorporating both anisotropy and remanent magnetization. For M_r

$=0$ or the dc field parallel to $\vec{H}_c$, the anisotropy parameter deduced from our data may be quite accurately described as $K(T)=K(0)(1-\beta T/T_g')$; where T_g' is at the peak of the magnetization measured at ≈ 3 kG. We find $K(0)$ may be expressed as $aC_{\mathrm{Mn}}{}^2+b_iC_{\mathrm{Mn}}C_i$. The dependence of the field for resonance H_r as a function of H_c and θ is anomalous. Specifically, H_r is very anisotropic while the total magnetization of the sample, as measured simultaneously, remains isotropic to within the experimental accuracy of a few percent. We suggest that this requires the introduction of another "memory" variable, not in our present model, which only exists below T_g'. This variable may be crucial to an understanding of the nature of the anisotropy fields in the spin-glass state.

In Fig. 1, we present the value of H_r as a function of spectrometer frequency for $C_{\mathrm{Mn}}=10$ at.%, at $T=1.8$ K and $H_c=0$. Under these conditions the magnetization, M, is virtually all paramagnetic. The data are best represented by a linear relationship of the form $\omega/\gamma=\alpha H_r+H_i$. We note that the slope is far from unity, and within the error may be taken as $+\frac{1}{2}$. Monod and Berthier[2] have performed analogous low-frequency ESR measurements using predominantly remanent magnetization. They also found a linear relation, but

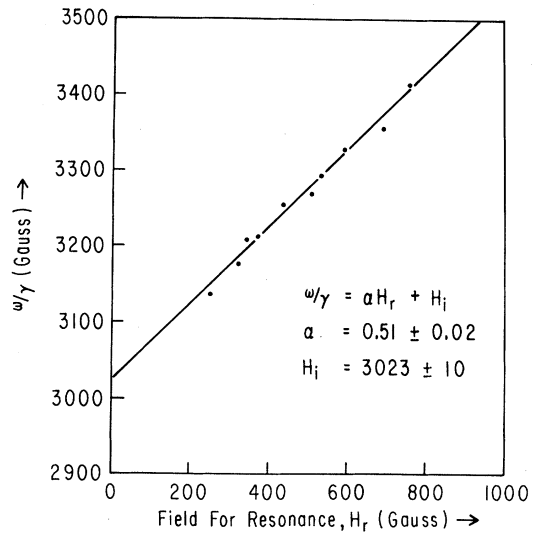

FIG. 1. The field for resonance H_r (in gauss) vs the spectrometer radian frequency ω/γ(G) for a CuMn alloy ($C_{\mathrm{Mn}}=10$ at.%) at 1.8 K. [The sample was cooled in zero field ($H_c=0$).] The data are best represented by a linear relation whose slope is essentially $\frac{1}{2}$. The slope and intercept, H_i (in gauss), are compared with a theoretical model as discussed in the text.

with different values of slope and intercept. From both of these data it is now clear that the antiferromagnetic resonance relation originally proposed in the pioneering paper of Owen, Brown, Arp, and Kip[3] does not apply whether the magnetization is either paramagnetic or remanent.

The simplest model free energy we could invent that incorporates magnetic remanence, anisotropy, and Zeeman energy is

$$F=(2\chi_\perp)^{-1}(\vec{M}-M_r\hat{n})^2+\frac{1}{2}\left(\frac{1}{\chi_\parallel}-\frac{1}{\chi_\perp}\right)(\vec{M}\cdot\hat{n}-M_r)^2 - \tfrac{1}{2}K(\hat{N}\cdot\hat{n})^2-\vec{M}\cdot\vec{H}, \quad (1)$$

where $\hat{n}$ is a direction associated with the order parameter, and $\hat{N}$ is a fixed (in space) direction whose origin remains a mystery to us; $\chi_\perp$ and $\chi_\parallel$ are the principal values of the susceptibility tensor.[4] We derive equations of motion for $\vec{M}$ and $\hat{n}$, neglecting dissipation, by forming Poisson brackets with F, using the symmetry-derived relations $[M_i,M_j]=\gamma\epsilon_{ijk}M_k$, $[M_i,n_j]=\gamma\epsilon_{ijk}n_k$.[5] We find

$$d\vec{M}/dt=\gamma\vec{M}\times\vec{H}-\gamma K(\hat{N}\cdot\hat{n})(\hat{N}\times\hat{n}),$$
$$d\hat{n}/dt=\gamma(\vec{M}/\chi_\perp-\vec{H})\times\hat{n}. \quad (2)$$

For static $\vec{H}$ parallel to $\hat{N}$ and not too big, $\hat{n}=\hat{N}$ and $\vec{M}=(M_r+\chi_\parallel H)\hat{N}$ yields the minimum of the free energy. Linearizing the equations of motion about this equilibrium yields resonant frequencies

$$\frac{\omega^\pm}{\gamma}=\pm\left(\frac{1+\xi}{2}H-\frac{M_r}{2\chi_\perp}\right) +\left[\left(\frac{1-\xi}{2}H+\frac{M_r}{2\chi_\perp}\right)^2+\frac{K}{\chi_\perp}\right]^{1/2}, \quad (3)$$

where $\xi=1-\chi_\parallel/\chi_\perp$.[6] For $M_r=0$, $H_i=(K/\chi_\perp)^{1/2}$, and if $\frac{1}{2}(1-\xi)H\ll H_i$, then $\omega^\pm/\gamma=\pm\frac{1}{2}(1+\xi)H+H_i$, so that $\alpha=\frac{1}{2}(1+\xi)$.

Since the slope in Fig. 1 is $\frac{1}{2}$, we assume that $\chi_\parallel=\chi_\perp$, i.e., $\xi=0$, in all the theoretical analysis that follows.

In Fig. 2, we present a plot of Eq. (3) for $M_r=0$. We believe that all the zero-field–cooled ESR data in the literature for CuMn are consistent with this representation. We are immediately led to examine three important questions: (1) Does the second mode (ω^-) exist, and if so, what are its properties? (2) If we assume some model validity, what is the behavior of the anisotropy parameter K as a function of T, C_{Mn}, and C_i with $H_c=0$? (3) How well does the model represent the data when $M_r\neq 0$? We address these questions in turn.

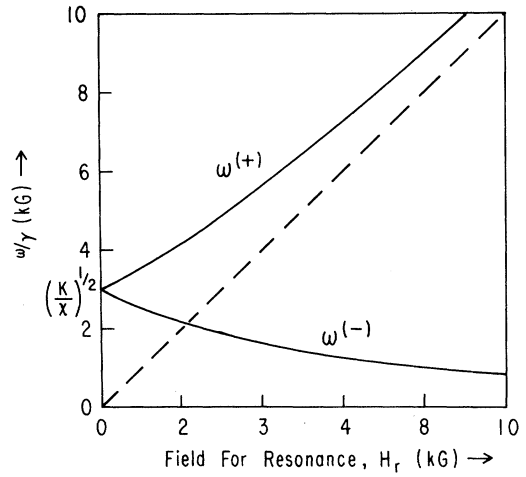

FIG. 2. The field for resonance H_r vs (radian frequency)/γ(kG) corresponding to the two resonance modes as represented in Eq. (3) for zero remanent magnetization ($M_r = 0$) and $\chi_{\parallel} = \chi_{\perp}$. The intercept is given by $(K/\chi_{\perp})^{1/2}$. The ω^+ mode is asymptotic to the dashed line $\omega = \gamma H_r$.

In Fig. 3, we present traces representing the observation of both ESR modes in the same sample. ESR data are normally taken at a fixed spectrometer frequency with the magnetic field being swept through the resonance condition. The sample temperature may be conveniently changed. In these alloys, H_r decreases from values corresponding to ω/γ at high temperatures to much smaller values at low T. This behavior may be conveniently visualized by reference to Fig. 2 and assigning an increase to H_i as T is reduced. If one starts off seeing the usual (ω^+) ESR mode at higher temperatures, one cannot see the ω^- mode until H_r has been reduced to zero, and then the temperature has been lowered further. We initially prepared an alloy of $C_{Mn} = 15$ at.% to allow us to reach this condition at convenient temperatures ($\simeq 20$ K). While it was possible to observe the ESR signal as H_r went to zero, we did not see the onset of another mode. However, we have also been studying the reduction of H_r (more usually referred to as an enhancement of the shift $\delta H = \omega/\gamma - H_r$) as a function of added impurities, following up on the important experiments of Okuda and Date.[7] In a CuMnNi sample ($C_{Mn} = 8$ at.%, $C_{Ni} = 0.3$ at.%) we indeed found the second mode as shown in Fig. 3(b). Ni was chosen as it had the largest lineshift per broadening of the

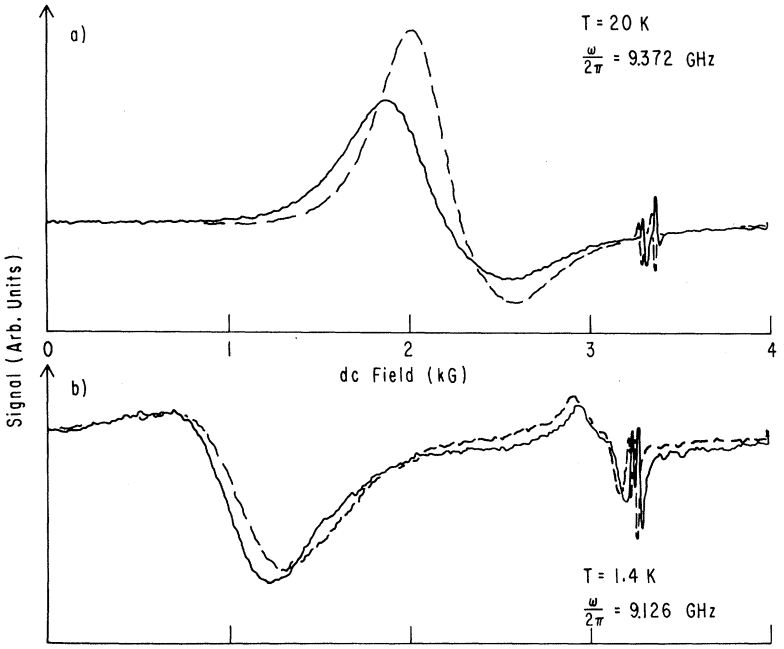

FIG. 3. Traces of the spectrometer ESR signal vs the dc field for a CuMnNi sample ($C_{Mn} = 8$ at.%, $C_{Ni} = 0.3$ at.%). The solid curves are for increasing field, and the dashed curves for subsequent decreasing. The structure at 3 kG is due to DPPH magnetization measuring markers (Ref. 1). (a) Choice of temperature and frequency for which only the usual ω^+ mode is observed. (b) Choice of temperature and frequency for which the new ω^- mode is observed. The special phase relationship between the ω^+ and ω^- modes is contained in the model (Ref. 9).

elements studied in Ref. 7. We have not yet characterized this new mode as thoroughly as the usual mode, but find the following features as predicted by the theory: (a) At fixed frequency, H_r shifts to higher values with decreasing temperature or with application of a small H_c. (b) The ω/γ vs H_r relation is linear with a slope of value -0.4 ± 0.04.[8] (c) The lineshape is phase shifted relative to the usual mode.[9]

For the ω^+ mode, Eq. (3) may be solved for K and expressed in a particularly simple form; $K = M^0 \delta H$, where M^0 is the total magnetization at the field ω/γ. As mentioned,[1] we have been able to make *in situ* measurements of the total magnetization under all sample conditions using the field separation between a pair of suitably prepared and placed diphenylpicrylhydrazyl (DPPH) spin markers. The g value of DPPH is very close to 2.0, so that our magnetization measurement corresponds to precisely the quantity M^0. For δH and M^0 data taken under $H_c = 0$ conditions, we deduce the temperature dependence of K, and find surprisingly straight lines of the form $K = K(0)(1 - mT)$, where m is a constant that depends on C_{Mn}. From the dependence of $K(0)$ as just defined upon sample parameters, we find $K(0) = aC_{\mathrm{Mn}}^2 + b_i C_{\mathrm{Mn}} C_i$ G^2 with $a = 115 \pm 5$ and $b_i = 3050 \pm 300$ for nickel. The quadratic dependence on concentration suggests pairwise interactions. When we plot $K(T)/K(0)$ vs T/T_g' we find for the concentrations studied ($C_{\mathrm{Mn}} = 2$, 4, 8, and 10 at.%), $K(T) = K(0)(1 - \beta T/T_g')$, where $\beta = 0.67 \pm 0.07$.[10]

With respect to our question (3), we have analyzed the data presented in Ref. 2, utilizing our best estimate of their remanent magnetization under the conditions described. We find satisfactory agreement both for values of K, from their values of ω_a, and for their slopes (which differ slightly, but significantly from unity).[11] We find that for our data taken under conditions of H_c up to 3 kG ($\simeq \omega/\gamma$) there is a mild reduction in $K(T)$ of $\simeq 10\%$.

While all of the data just discussed appear to be interpretable within the framework of our model, there is one new class of data which is not, and which we feel is worthy of detailed attention. In Fig. 4 we present data for H_r as a function of θ, for several values of H_c. We have taken such data on numerous other samples including those doped with Ni. Space prohibits a detailed description of all the types of angular dependence observed, but it is clear that the samples have a very anisotropic behavior. (In one sample for example, at $\theta = 180°$, H_r exceeded ω/γ.) The observation we find most important is that the simultaneous magnetization measurements of M^0 by the procedures described imply that M^0 is constant in magnitude and simply rotates with the applied field over the full 360°. For example, for the sample of Fig. 4 with $H_c = 3$ kG, this constancy is to within our experimental accuracy of $\pm 2\%$. The anisotropic behavior is also manifested in the linewidths, but all the ESR signal characteristics smoothly return to the zero-field–cooled values at T_g'. Thus, we conclude that there is another direction or memory parameter which plays a significant role in determining the angular anisotropy needed to completely specify the spin-glass state, and that this parameter reduces to 0 at T_g'.

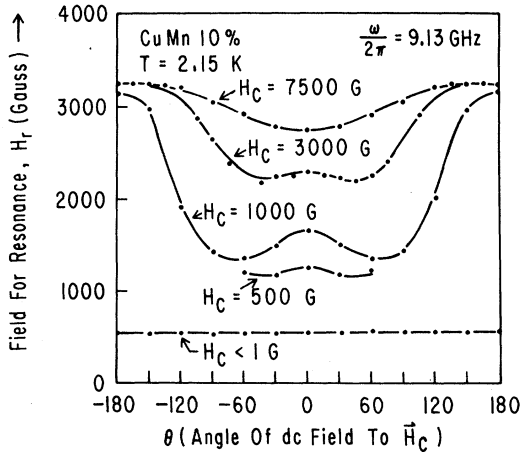

FIG. 4. Field for resonance H_r vs θ, the angle of orientation of the dc field to the direction of $\vec{H}_c$, for several values of H_c. Despite the marked anisotropy of $\vec{H}_r$ for $H_c > 1000$ G, in all cases the magnetization (measured at $\simeq 3$ kG) is isotropic within the experimental accuracy of a few per cent.

We gratefully acknowledge discussions with Professor J. Wilkins, who stimulated our interest in making ESR measurements in the spin-glass state. We thank Professor Shang-keng Ma and Professor R. Orbach for helpful conversations. We thank Professor W. M. Saslow for a stimulating preprint.[12] We thank Mr. Roger Isaacson for his generous help with the experimental apparatus. We wish to express our appreciation to the National Science Foundation for their continuing support under Grant No. DMR-77-22781-A02.

[1]The ESR measurements were performed in a conventional manner. The magnetization measurements were

made *in situ* on the sample in the ESR cavity. We observe the separation between the ESR signals from a pair of thin DPPH spin markers placed 90° apart on the central plane of a cylindrical sample. The details of the technique, computer simulations, experimental checks via a Faraday susceptometer, etc., will be presented in a future publication. The samples were prepared in an arc furnace, annealed under argon at 1100 K for 24 h, spark cut into cylinders, and heavily etched. Both quenched and slowly cooled samples produced consistent results.

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[6]The two modes correspond to the two circular polarizations of $\delta\vec{M}$, with (+) corresponding to the customary polarization sense in magnetic resonance. $\omega^{-}>0$ for H such that $\hat{n}=\hat{N}$ is the stable equilibrium.

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[8]Should more accurate measurements confirm that the magnitude of the slope is less than $\frac{1}{2}$, it would imply that $\chi_{\parallel}>\chi_{\perp}$ in this alloy.

[9]Equation 2 can be used to derive $\chi'(\omega)$, the real part of the rf susceptibility for linearly polarized rf magnetic field. Recall that the dc field H is varied; the singular part of $\chi'(\omega)$ turns out to be $(\chi\omega/2r)(H-H_r)^{-1}$ for $\omega/\gamma>H_i$, and $(\chi\omega/2r)(H_r-H)^{-1}$ for $\omega/\gamma<H_i$, where H_r is the field for resonance: $\gamma H_r=|\omega^2-(\gamma H_i)^2|/\omega$.

[10]Note that when measured in fields $\simeq 3$ kG, $T_g'\simeq 0.9T_g$ (T_g determined at low fields is given by $9.5\,C_{\mathrm{Mn}}{}^{0.65}$). We use T_g' in our relation because experimentally it is also the temperature where the ESR anisotropy effects (as measured in fields = 3 kG) have gone to zero. We also find that with the addition of 0.3 at.% Ni to CuMn alloys there is no change in T_g' to within ± 5%.

[11]Values of K determined from hysteresis-loop areas agree with our scaling but are smaller as would be expected. P. Monod, J. Prejean, and B. Tissier, J. Appl. Phys. 50, 7324 (1979). Values of K deduced from zero-field NMR agree with our values. H. Alloul, J. Appl. Phys. 50, 7330 (1979).

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ESR Study of the Kondo Effect in *Au*:Yb

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Paramagnetic resonance experiments at 3 and 9 GHz were performed in dilute *Au*:Yb in the temperature range from 100 mK to 4 K. All results show for the first time a logarithmic temperature dependence for the g-value shift and for the relaxation rate in complete agreement with theoretical predictions. The fit yields a Kondo temperatue of $T_K\sim 10$ μK and a degeneracy of $d\sim 3$ for the local-moment–conduction-electron interaction channels.

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It has been pointed out by Orbach[1] that the ESR of local moments could, in principle, be used to observe the Kondo effect directly. The change of the local susceptibility and the relaxation rate of local moment could be determined independently in one experiment. However, the ESR will be limited to a temperature range $T>T_K$, where the local moment is well defined. Most of the experiments in the field of Kondo effect in dilute alloys did measure the static properties (susceptibility, thermopower, etc.) or the scattering rate (T-matrix) via the resistivity anomaly. Only a few experiments determined the relaxation rate of the local moment, e.g., neutron scattering[2] and NMR[3] in *Cu*:Fe or Mössbauer effect[4] in *Au*:Yb.

The first observation of the ESR for the *Au*:Yb system was made by Hirst *et al.*,[5] where the g-value shift was shown to be negative, indicating that the interaction between the conduction electrons of the host and the Yb impurities is antiferromagnetic. In this Letter we present the first experimental evidence of the Kondo effect in the linewidth and the g shift in an ESR spectrum. We choose *Au*:Yb because the ESR of this system is not bottlenecked and the full effect on the exchange should be visible.

The commonly accepted description for a nonbottlenecked ESR is given by[6]: (1) g shift, $\Delta g=g_{\mathrm{metal}}-g_{\mathrm{ionic}}=\alpha N(E_F)J_1$, and (2) relaxation rate of the local moment to the conduction electrons, $\hbar\tau^{-1}=\hbar\delta_{ie}=\pi\alpha^2[N(E_F)J_2]^2kT$, where α is the modified de Genne factor $\alpha=(g_J-1)g_J{}^{-1}g_{\mathrm{eff}}=0.418$