Characteristic temperatures in the spin-glass *Au*Fe

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The binary-alloy system *Au*Fe with varying concentration, c, of Fe is shown to have three distinct regimes with quite different physical properties. These are the *Kondo* regime at low concentrations, the *spin-glass* regime at intermediate concentrations, and the *dilute-ferromagnet* regime at high concentrations. This paper contains a comprehensive compilation of experimental data expressed in terms of *characteristic temperatures*, like, for example, the spin-glass freezing temperature and the temperature of the resistance maximum. A coincidence of the characteristic temperatures is evident in the spin-glass regime $0.05\% \lesssim c \lesssim 15\%$. Here a recent theoretical calculation based on the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect-exchange interaction between the Fe spins accounts quantitatively for the observed dependence on concentration and electronic mean free path in a variety of different experiments. This includes, for example, recent experiments in quench-condensed films showing a large mean-free-path effect on the resistance maximum. The agreement extends to remarkably high concentrations and demonstrates substantial self-damping of the RKKY interaction. At the transition into the dilute ferromagnet regime $c \gtrsim 15\%$, near the threshold for nearest-neighbor percolation, there is a *sharp* departure of the observed freezing temperatures away from the predicted concentration dependence. This indicates a change-over between the spin-glass state dominated by the RKKY interactions and the dilute ferromagnet state dominated by nearest-neighbor direct-exchange interactions and is interpreted qualitatively in terms of recent percolation theories. Entering the Kondo regime $c \lesssim 0.05\%$ leads to a dispersion of the characteristic temperatures, whereby the complete agreement with the theoretical calculation is lost. Only one of the characteristic temperatures, the noise temperature derived from the resistance maximum by a method which incorporates the Kondo effect, remains in agreement with the RKKY calculation.

I. INTRODUCTION

The question of sharp versus nonsharp features in the observed properties of spin-glasses is currently of considerable interest.[1,2] An example of a sharp feature is the cusp in the low-field ac magnetic susceptibility, while a nonsharp feature is the maximum in the electrical resistivity, both properties considered as functions of the temperature. A sharp feature is likely to involve a large number of correlated degrees of freedom, which in spin glasses are the spins on the randomly distributed magnetic atoms in the metallic alloy. Accordingly, the cusp in the susceptibility at the temperature T_0 is considered to signal the onset of a macroscopic freezing of the spins into fixed, but more or less random directions. The precise nature of this phenomenon is not understood at present.[2]

On the other hand, a nonsharp feature may not require the same amount of correlation between the degrees of freedom concerned. In fact, it was recently shown[3,4] that the resistivity maximum may be explained by the scattering of the conduction electrons against individual spins in the presence of noise generated by their interactions already at temperatures well above the freezing temperature. Hence in the process of cooling the effects of the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect-exchange interaction[5] are clearly observable before they lead to the freezing at T_0. Therefore, a complete understanding of the properties of spin-glasses encompasses both the sharp and the nonsharp features produced by the interactions. In particular it must be demonstrated *quantitatively* that such different characteristics do in fact originate in the same interaction.

In the present paper I show that this is indeed the case for a comprehensive selection of data on the spin-glass *Au*Fe. A corresponding analysis of the system *Cu*Mn will be published in a separate paper.[6] In these studies a range of fractional concentrations of Fe and Mn covering some five or six decades will be considered. This includes all compositions of the Au- and Cu-rich alloys that have been investigated over the years.

In the course of this analysis some interesting additional information is obtained. It is possible to demonstrate that dominance of the RKKY interaction, perhaps surprisingly, extends to rather high concentrations. Only at the nearest-neighbor percolation thres-

hold around 15–20 at.% of magnetic atoms does another interaction, which is presumably direct exchange, start to control the freezing process. The evidence for this in *Au*Fe will be discussed at length in the following. It is found that damping of the RKKY interaction, due to the finite mean free path of the conduction electrons that mediate it, plays an important part at the higher concentrations. A quantitative evaluation of this effect is possible by the correlation of observed freezing temperatures and observed resistivities, and will be carried out within the framework of a recent theoretical study by the present author.[7]

The importance at higher concentrations of the interplay between long-range RKKY and short-range direct-exchange interactions was emphasized by Coles.[8] Recently, on the basis of magnetization[9,10] and neutron-scattering[11–14] experiments in the 10–20-at.% range, Murani suggested that spin-glass characteristics persist, even in the presence of direct exchange, all the way up to the percolation threshold for dilute ferromagnetism at about 17 at.%. The present analysis provides strong quantitative support of this viewpoint. Interestingly, Borg and Dienes[15] found a strong effect of short-range ordering induced by annealing or neutron irradiation in a 17.5-at.% sample, while there was no such effect in 13.5- and 24-at.% alloys. Very recently, Maartense and Williams[16] found a complicated behavior in a 22-at.% alloy, which is a dilute ferromagnet.

Within the framework of the Edwards and Anderson spin-glass model[17] Sherrington and Southern[18] discussed the existence of spin-glass and ferromagnetic phases for different ratios of mean value to width of the distribution of exchange couplings. Work along this line has been continued.[2,19] At present it is not clear to what extent this model applies to the situation in real spin-glasses.

It is known how the RKKY interaction varies with pressure from studies of the resistivity.[3,4,20–26] It is an attractive prospect that the region around the percolation threshold may be investigated by studying the pressure dependence of the freezing temperature. This is discussed in more detail in Sec. V, where a quantitative prediction of the expected variation is given.

The results communicated in the present paper are also relevant for systems of rare-earth spin-glasses. It is shown that even in the absence of direct exchange there will be significant effects due to the damping of the RKKY interaction. Furthermore, due to their relatively larger resistivitites, amorphous spin-glasses can be expected to have large damping effects. These will have to be taken into account in the discussion of their properties, preferably through information acquired by resistivity measurements on the same or identical samples. In quench-condensed films with very large resistivities a substantial variation of the resistivity maximum with annealing temperature was found by Korn[27] and recently studied in more detail by Buchmann *et al.*[28] This effect will be quantitatively accounted for in Sec. IV.

The observed concentration dependences are highly nontrivial. In particular, the characteristic temperatures are not exactly proportional to the concentration, although they do increase with increasing concentration. Therefore, identifying an RKKY spin-glass by looking for scaling laws[29] that require *strict* proportionality tends to exclude even many of the canonical spin-glass alloys. However, one can show that two separate effects are responsible for this deviation from linearity when the RKKY interaction is the relevant interaction. These are the damping[30] referred to above and fluctuations in local concentration[31] distinguishing a truly random distribution of spins from a uniform distribution. Including these two effects[7] it is possible to account in a quantitative way for the observed concentration dependence over the full range where the RKKY interaction remains the basic coupling. In fact, Souletie[32] has shown that mean-free-path effects can explain some of the deviations from perfect scaling at higher concentrations in the specific heat of *Au*Fe (and other systems and properties as well). The conclusion reached here is that this is necessary but not sufficient, and that concentration fluctuations must be included as well to obtain a quantitatively correct theory. In relation to this it is interesting to note that Liu and Smith[33] have recently shown that by scaling the temperature with the observed characteristic temperature, rather than with the bare concentration, the scaling in the susceptibility of *Au*Fe may in fact be considerably improved. This will simultaneously improve the scaling in the specific heat, as was originally demonstrated by Dreyfus *et al.*[34] It appears that this principle is of general validity.

At low concentrations Tholence and Tournier[35–37] and most recently Frossati *et al.*[38,39] found a decrease of the freezing temperature with decreasing concentration, below the prediction of the scaling laws. This pattern is not substantially changed by the modifications of the concentration dependence referred to above. The effect was correlated with the emergence of the Kondo effect. This interpretation is supported by the present investigation including other properties of *Au*Fe that also show anomalous behavior in the same region, and particularly by the analogous investigation[6] of *Cu*Mn that shows similar anomalies to be absent until much lower concentrations, in accordance with the smaller Kondo temperature in *Cu*Mn. Interesting new aspects of the spin-glass–Kondo transition are discussed in a separate paper.[3]

II. DATA

The amount of data on the classical spin glass *Au*Fe is very large. Usually measurements where tempera-

ture, magnetic field, pressure, etc. are changed in an essentially continuous fashion are available. Todays theories do not provide fully adequate means of handling this large diversity of information. Hence in the present paper a much less ambitious approach is adopted based on the analysis essentially of the *magnitude* of the effect in each experiment. This is given in terms of a suitably defined *characteristic temperature*. In the cases where the magnitudes are characteristic fields for example, these are converted into equivalent temperatures.

The selection of characteristic temperatures includes T_0, $T_{\ddot{0}}$, T_H, T_m, and Δ_c, defined in the following ways: (a) The magnetic susceptibility in low static or alternating fields has a cusp or rounded maximum at the temperature T_0, usually called the freezing temperature. (b) Hyperfine splitting appears in Mössbauer spectra on cooling below the temperature $T_{\ddot{0}}$ (precise definitions are given by the various investigators). (c) The temperature T_H is extracted from the high-field magnetization according to

$$M \simeq M_{\text{sat}}(1 - H_0/H) \ , \tag{1}$$

where M is the magnetization, M_{sat} is the saturation magnetization, and H is the magnetic field. For low temperatures[33] T one has

$$H_0 \simeq (1.185 T_H + A'T) k_B/g\mu_B \ . \tag{2}$$

Here k_B is Boltzmann's constant, μ_B is the Bohr magneton, $g=2$, and A' is a constant. The constant number in front of T_H has been chosen such as to make T_H coincide with T_0 in *both* *Au*Fe and *Cu*Mn (it is not obvious that this is possible with the same constant, but it indicates that T_H and T_0 may have similar dependences on the magnitude of the spin S and other parameters distinguishing the two systems). (d) The excess resistivity

$$\Delta\rho = \rho - \rho_{\text{Au}} \ , \tag{3}$$

where ρ is the total resistivity of the alloy and ρ_{Au} is the resistivity of pure gold, has a maximum at the temperature T_m. (e) The noise temperature Δ_c is calculated from T_m of the resistivity and the Kondo temperature T_K according to (79) of Ref. 3,

$$\Delta_c = T_m Q(\ln(T_m/T_K)) \ , \tag{4}$$

where

$$Q(x) = \frac{2(R_2 + R_1 x + x^2)}{i_0(R_5 + R_4 x + R_3 x^2 + x^3)} \ , \tag{5}$$

$i_0 = 1.086$, $R_1 = 9.527$, $R_2 = 20.792$, $R_3 = 10.000$, $R_4 = 25.375$, and $R_5 = 24.610$.

This selection includes mainly properties where data for many different concentrations are available.[40] Well-defined characteristic temperatures may also be obtained from other properties, like neutron-scattering

TABLE I. Freezing temperatures T_0 derived from susceptibility or magnetization. Data points in Fig. 1: Rounded maximum (○), and cusp (⊙).

c (at.%)	T_0 (K)	Source	Reference
37	503	Pan, Kaufmann, and Bitter	41
0.5	4	Lutes and Schmit	42
1.0	7.4		
8	25	Béthoux, Ishikawa, Souletie, Tournier, and Weil	43
25	283	Sundahl, Sivertsen, and Chen	44
14.9	109	Crangle and Scott	45
19.5	201		
24.2	296		
29.5	381		
0.1	2.1	Cannella, Mydosh *et al.*	46–52
0.2	3.0		
0.5	5.4		
0.91	8.5		
1.0	9.0		
2.0	13.9		
5.0	22.2		
7.6	27.9		
12	36		
13	38		
0.0208	0.15	Tholence, Tournier *et al.*	35–37
0.0283	0.24		
0.0358	0.36		
0.0570	0.58		
0.1	1.2		
0.2	2.4		
0.5	5.2		
1	8		
3	15		
8	28		
15	50	Murani	9
17	130	Mydosh, Ford, Kawatra, and Whall	58
22	220		
1	8.3	Werner	53
8	29		
0.25	2.7	Guy	54–56
2	13.7		
4	20.6		
7	27.7		
0.605	6.05	Liu and Smith	33
1.5	11.6	Murnick, Fiory, and Kossler	57
0.0034	0.0052	Frossati, Tholence, Thoulouze, and Tournier	38–39
0.0059	0.0195		
0.0083	0.042		
0.0108	0.071		
22	213.1	Maartense and Williams	16
2.1	12	Domb, Sellmyer, Quick, and Borg	40

intensity,[11–14] specific heat,[29,32,34,53,85–87] depolarizing fields in muon precession studies,[57] magnetoresistance,[68,80,88–90,107] anomalous Hall effect,[91–100] thermal conductivity,[77] thermopower,[70,101–110] and others.[111] The collection of data given in the tables and figures is not claimed to be exhaustive, although it is rather comprehensive. Some hitherto unpublished data have been included. No attempt was made to evaluate the quality of the data, and early data have been included alongside more recent. The sources are indicated in the tables.

The prime difficulty in many of the cases is well illustrated by the maximum in the resistivity, namely, that even when a characteristic temperature (T_m) is readily obtained from the measurements, it is only by the application of a theoretical decoding that the quantity holding the information about the RKKY interactions in a "clean" form (Δ_c) appears. Fortunately, there are cases like the cusp in the susceptibility (T_0) where this is not a serious problem over a considerable fraction of the concentration range. Furthermore, the freezing temperature $T_{\ddot{0}}$ deduced from the Mössbauer effect studies as given by the authors are extracted from the spectra in different ways. One is referred to the original papers for these details, as they do not appear to influence the deduced $T_{\ddot{0}}$ above the level of statistical uncertainty. In some early resistivity measurements values of T_m are not listed by the authors and had to be extracted from the published graphs with the ensuing uncertainty. Sometimes only the total resistivity was given and the resistivity of gold had to be subtracted before T_m was determined. This is required for consistency as the temperature-dependent phonon resistivity of pure gold could influence the value of T_m. In some of the most important cases the original data were made available for analysis, however. Deviations from Matthiessen's rule[117] remain an important source of systematic error in the analysis of $\Delta\rho$ when the concentration is large enough that T_m is bigger than about 10 K. Presumably, therefore, the data shown in Fig. 1 at these concentrations cannot be analyzed by the theory of the resistivity in Ref. 3 in a meaningful way. This is apparent in the lack of concentration dependence of T_m, and accordingly in the deduced value of Δ_c. These data are shown for completeness but not further considered in the following analysis.[118]

It is obvious from Figs. 1 and 2 that a certain amount of scatter is present in the data. This is due to a variety of causes. Most important besides those mentioned above is presumably differences in preparation of the alloys by different investigators, including some uncertainty in the concentration, whether nominal or analyzed. On the other hand, even in the presence of these uncertainties the data are consistent enough for the analysis of the present paper to be meaningful, even to the extent of extracting numerical values of model parameters to be introduced in the next section. This is mainly due to the fairly large amount of data that exist for *Au*Fe. The situation is more difficult with most other spin-glass systems at the moment, with the exception of *Cu*Mn. It is a potential benefit of a study of the present nature, that it

TABLE II. Freezing temperatures $T_{\ddot{0}}$ derived from Mössbauer effect measurements. Data points in Fig. 1: (×).

c (at.%)	$T_{\ddot{0}}$ (k)	Source	Reference
0.84	7	Borg, Booth, and Violet	59
1.85	12		
7.38	28		
10.2	31		
11.5	35		
3.3	19	Gonser, Grant, Meechan, Muir, Jr., and Wiedersich	60
6.3	28		
15.7	54		
19.5	165		
24.7	303		
5	23	Craig and Steyert	61
0.84	11	Violet and Borg	62
1.7	14.8		
1.9	16		
2.9	17		
4.4	23.5		
6.7	27.6		
7.4	27.8		
8.0	31	Violet and Borg	63
10.5	40		
12.8	32	Ridout	64
5.0	25	Borg and Pipkorn	65
9.0	37		
12.0	38		
15.0	45		
2.0	17	Longworth	40
2.8	18	Quick, Borg, Pipkorn, and Violet	40
17.5	55	Borg, Lai, and Violet	66
13.5	45	Borg and Dienes	15
24.0	280		
2.6	18	Domb, Sellmyer, Quick, and Borg	40

TABLE III. Characteristic temperature T_H derived from high-field magnetization. Data points in Fig. 1: (⊘)

c (at.%)	T_H (K)	Source	Reference
0.0042	0.41	Liu and Smith	33
0.0094	0.56		
0.0169	0.56		
0.0242	0.81		
0.0572	0.91		
0.1160	1.42		
0.2225	2.36		
0.6050	6.18		

may help to identify the parameters in the preparation of alloys that influence their properties. A step in that direction is taken by the analysis of the influence of annealing temperature on the properties of quench-condensed films, which are discussed in more detail in the following.

Also given in Table IV and shown in the top section of Fig. 1 are values of the *total* resistivity divided by the absolute fractional concentration c. Of relevance for the evaluation of the damping of the RKKY interaction are the resistivities at the temperatures T_0 (for the evaluation of T_0) and T_m (for the evaluation of Δ_c). At the concentrations where $\rho(T_m)$ is available it coincides rather well with $\rho(T_0)$ due to the relatively small temperature dependent content and to the closeness of T_0 and T_m. These values are subject to some additional uncertainty besides what was listed above. Thus, for example, temperature-independent defect scattering and length-to-area uncertainties affect the absolute value of the resistivity, but not the position of the maximum, hence not T_m. On the other hand, T_0 and Δ_c are not very sensitive to these uncertainties, as changes of the order of 20% in ρ are needed to change the calculated freezing temperature by an amount comparable to its uncertainty. The most significant feature of these data is that a very clear decrease in $\rho(T_0)/c$ by a factor of 2 takes place between 5- and 20-at.% Fe.

Evident in Figs. 1 and 2 is a rather good coincidence of characteristic temperatures, with the exception of T_m, over a range of concentrations extending upwards from 0.05%, thus covering nearly three decades. This is quite remarkable considering the rather complicated functional relationship emerging. The immediate conclusion is that in this range these different characteristic temperatures measure the same quantity, namely the strength of the RKKY interaction. Only for T_m is substantial theoretical processing required to extract this measure, i.e., Δ_c. There exists strong evidence from the analysis of the pressure dependence of T_m in a wide range of systems, including *Ag*Mn, *Au*Mn, *Cu*Mn, *Au*Cr, *La*Ce, *Au*Fe, *Mo*Fe, and *Cu*Cr,[3,4,22–26] that as long as $\Delta_c >> T_K$ (4) is a reliable decoding of the composite quantity T_m, that also depends on T_K, into the more fundamental Δ_c. The coincidence of Δ_c with the other measures of the interaction strength was therefore expected, but is nevertheless striking. Particularly so because it is sensitive to the value of T_K, and because the value $T_K = 0.19$ K is known with rather high accuracy from independent analysis—a fact that distinguishes *Au*Fe from most other spin glass and Kondo systems.

Note also in Fig. 1 at the lowest concentrations below 0.05% how *simultaneously* T_0 and T_H begin to deviate in opposite directions away from the smoothly decreasing Δ_c. The obvious interpretation of this is that these characteristic temperatures are showing an influence of the Kondo effect when T_K is approached. Here they must then be considered composite in a similar way as T_m, while Δ_c is *not* and remains a clean measure of the RKKY interaction strength. This interpretation is supported by the results of the comparison with the theoretical calculation to be described in the following section.

To summarize briefly this section, the observed coincidence of characteristic temperatures derived from sharp (T_0 and T_0') and nonsharp (Δ_c and T_H) features of the measured spin-glass properties is evidence for the *common origin* of the effects. This is generally recognized to be the RKKY interaction, as will be verified in the following.

III. THEORY

The essence of a recent calculation[7] of the freezing temperature is that two effects influence its dependence on the concentration c. These are local fluctuations in concentration due to the random distribution of spins and exponential damping of the RKKY interaction beyond the electron mean free path λ. The resulting expression is[7]

$$k_B T_0 = Ac\left[\frac{3}{c}\int_1^\infty \frac{dx}{x^4} e^{-rcx}(1 - e^{c'(1-x^3)})\right]^{1/2} , \quad (6)$$

where

$$\begin{aligned} A &= b_S J^2 (2l+1)^2 / 4E_F \ , \\ b_S &= [(2S+1)^4 - 1]^{1/2}/12 \ , \\ r &= 4\beta e^2 \bar{\rho}/a_0 h \ , \\ c' &= c/(1-c) \ , \quad \bar{\rho} = \rho/c \ , \end{aligned} \quad (7)$$

and where e is the electronic charge, h is Planck's constant, a_0 the fcc lattice constant, E_F the Fermi energy, $l = 2$ for $3d$ impurities, J the s-d exchange coupling constant, and β a correction factor to be discussed below.

The expression for the freezing temperature [(11) or (16) below] that leads to the result (6) is very general. In order to be specific, here as well as in Ref. 7 it was taken from the model of Edwards and Anderson,[17] whereby the value of b_S given in (7) is obtained.[18,124] The details of this rather simple calculation were given in Ref. 7 and will not be repeated here. However, before proceeding it is useful to make a few observations of a more general nature.

The quantity that appears on the right-hand side in (6) may be interpreted as the natural energy scale corresponding to the RKKY Hamiltonian[5]

$$\mathcal{H}_{\mathrm{RKKY}} = -\frac{1}{2}\sum_{i \neq j} \mathcal{J}(R_{ij}) \vec{S}_i \cdot \vec{S}_j \ , \quad (8)$$

where $\mathcal{J}(R_{ij})$ is the coupling of two spins at the dis-

TABLE IV. Characteristic temperatures T_m and Δ_c derived from resistivity measurements using $T_K = 0.19$ K. Also given are total resistivities $\rho(T_m)/c$ and $\rho(T_0)/c$ at T_m and T_0 (interpolated in Table I), divided by the concentration. Data points in Fig. 1: T_m (●), Δ_c (Δ), $\rho(T_m)/c$ (+), and $\rho(T_0)/c$ (○) (top section).

c (at.%)	T_m (K)	Δ_c (K)	$\rho(T_m)/c$ ($\mu\Omega$ cm)	T_0 (K)	$\rho(T_0)/c$ ($\mu\Omega$ cm)	Source	Reference
0.12	3.2		917			Linde	67
0.15	3	1.58				Gerritsen	68
0.23	8	3.33					
0.42	12	4.59					
0.62	~150	~38	984			Domenicali and Christenson	69
0.91	~150	~38	912				
1.28	~150	~38	871				
2.0	~170	~42	825				
5.1	~180	~45	745				
0.1	2.5	1.38				MacDonald, Pearson, and Templeton	70
4.53				22	695	Sundahl, Chen, Sivertsen, and Sato	71
12.58				37	477		
24.9				318	402		
0.15	4.7					Star	72–74
0.04	0.69	0.580	994	0.57	994	Ford, Whall, Loram *et al.*	75–76
0.04	0.68	0.575	960				
0.06	1.23	0.841	942				
0.06	1.29	0.868	939				
0.07[a]	1.40	0.918	1005				
0.07	1.60	1.006	960				
0.08	1.72	1.058	889				
0.10	2.23	1.271	848	1.3	846		
0.15	4.28	2.06	876	1.8	871		
0.20	5.6	2.52	821	2.4	801		
0.25				2.9	839		
0.35				3.8	769		
0.056	1.1	0.78				Garbarino and Reynolds	77
0.091	2.0	1.18	844				
0.096	2.2	1.26	822				
0.106	3.2	1.65	833				
0.153	3.8	1.88					
0.0200	0.214	0.311	1051			Laborde and Radhakrishna	78–79
0.0275	0.362	0.402	894				
0.0300	0.418	0.434	928				
0.11	2	1.2	820			Berman and Kopp	80
0.19	7	3.0	800				
0.5				5.4	782	Mydosh, Ford, Kawatra, Whall, *et al.*	58, 81–84
0.8				7.5	746		
0.91	170	42.4	922	8.5	744		
1.06				9.0	725		
1.98	165	41.3	836	13.9	748		
2.15				14	724		
3.0	140	35.8					
4.0	165	41.3					
5.0	225	54.1	831	22	768		
6.5	~300	~69.6					
8.0				28	620		
12				36	502		

TABLE IV. *(Continued).*

c (at.%)	T_m (K)	Δ_c (K)	$\rho(T_m)/c$ ($\mu\Omega$ cm)	T_0 (K)	$\rho(T_0)/c$ ($\mu\Omega$ cm)	Source	Reference
17				130	388		
22				220	348		
0.13	3.48	1.76	860			Schilling, Crone, Ford, Methfessel, and Mydosh	20
0.10	2.38	1.33	859			Schilling, Ford, Larsen, and Mydosh	4

[a]Cold worked.

tance R_{ij}. In the absence of damping[5]

$$\mathcal{J}(R)=\frac{9\pi J^2(2I+1)^2}{E_F(2k_FR)^3}\left[\cos(2k_FR)-\frac{\sin(2k_FR)}{2k_FR}\right], \tag{9}$$

where k_F is the Fermi wave number. When the conduction electrons have a finite mean free path λ there appears an exponential factor $e^{-R/\lambda}$ and the oscillations are phase shifted.[30] Further details are only known in various approximations. Recent calculations were done by Kaneyoshi,[119] and by Takanaka and Yamamoto.[120]

Basically the energy scale is a constant times the root-mean-square RKKY interaction energy of one spin (i) due to all the others (j). This may be understood in a straightforward way as the outcome of an instantaneous random walk in three dimension of fields $\vec{H}_{(i)j}$ acting on spin (i) due to spins (j) (it is convenient to let the fields have the dimension of energy). In the absence of directional correlations between any pair of spins $\vec{S}_j$ and $\vec{S}_{j'}$, each step of the field vector, corresponding to spin (j), is in a random direction. Its length is sampled from an isotropic distribution $W_{(i)j}(\vec{H}_{(i)j})$. Hence the distribution $W_{(i)}(\vec{H}_{(i)})$ of the total field $\vec{H}_{(i)}$, where

$$\vec{H}_{(i)}=\sum_{j\neq i}\vec{H}_{(i)j} \tag{10}$$

will also be isotropic. The average size of the field $\vec{H}_{(i)}$ at some spin (i) must then be obtained from the square root of the second moment of $W_{(i)}$, as the first moment vanishes. In Appendix A it is shown that the second moment of $W_{(i)}$ is equal to the sum of the second moments of all the *different* $W_{(i)j}$.

In the simplest case where the magnitude of $\vec{H}_{(i)j}$ is known with certainty when the distance R_{ij} is given, say $\vec{H}_{(i)j}=|\mathcal{J}(R_{ij})|$, the distribution $W_{(i)j}$ is a δ function, and by (A12) its second moment is just $[\mathcal{J}(R_{ij})]^2$, whereby one can take

$$(k_BT_0)^2\propto\left\langle\sum_{j\neq i}\mathcal{J}(R_{ij})^2\right\rangle . \tag{11}$$

The $\langle\,\rangle$ means an average over spins (i). Hence k_BT_0 is proportional to the root-mean-square interaction energy.

TABLE V. Characteristic temperatures in quench-condensed films annealed at T_a. Using $T_K=0.19$ K.

c (at.%)	T_a (K)	T_m (K)	Δ_c (K)	ρ ($\mu\Omega$ cm)	r	Source	Reference
0.20	30	4.35	2.08	25.0	190	Korn	27
	90	4.25	2.04	21.3	162		
	190	4.6	2.17	14.0	107		
	290	5.6	2.52	9.62	73		
	380	7.0	3.00	4.41	34		
0.24	50	5.3	2.42	16.8	107	Buchmann, Falke, Jablonski, and Wassermann	28
	150	6.0	2.66	12.0	76		
	220	6.5	2.83	9.6	61		
	300	8.5	3.49	4.3	27		
0.6	9	13.5	5.05				
	50	14.5	5.35				
	120	14.8	5.44				
	220	16.8	6.03				
	300	21.0	7.23				
	470	23.0	7.79				

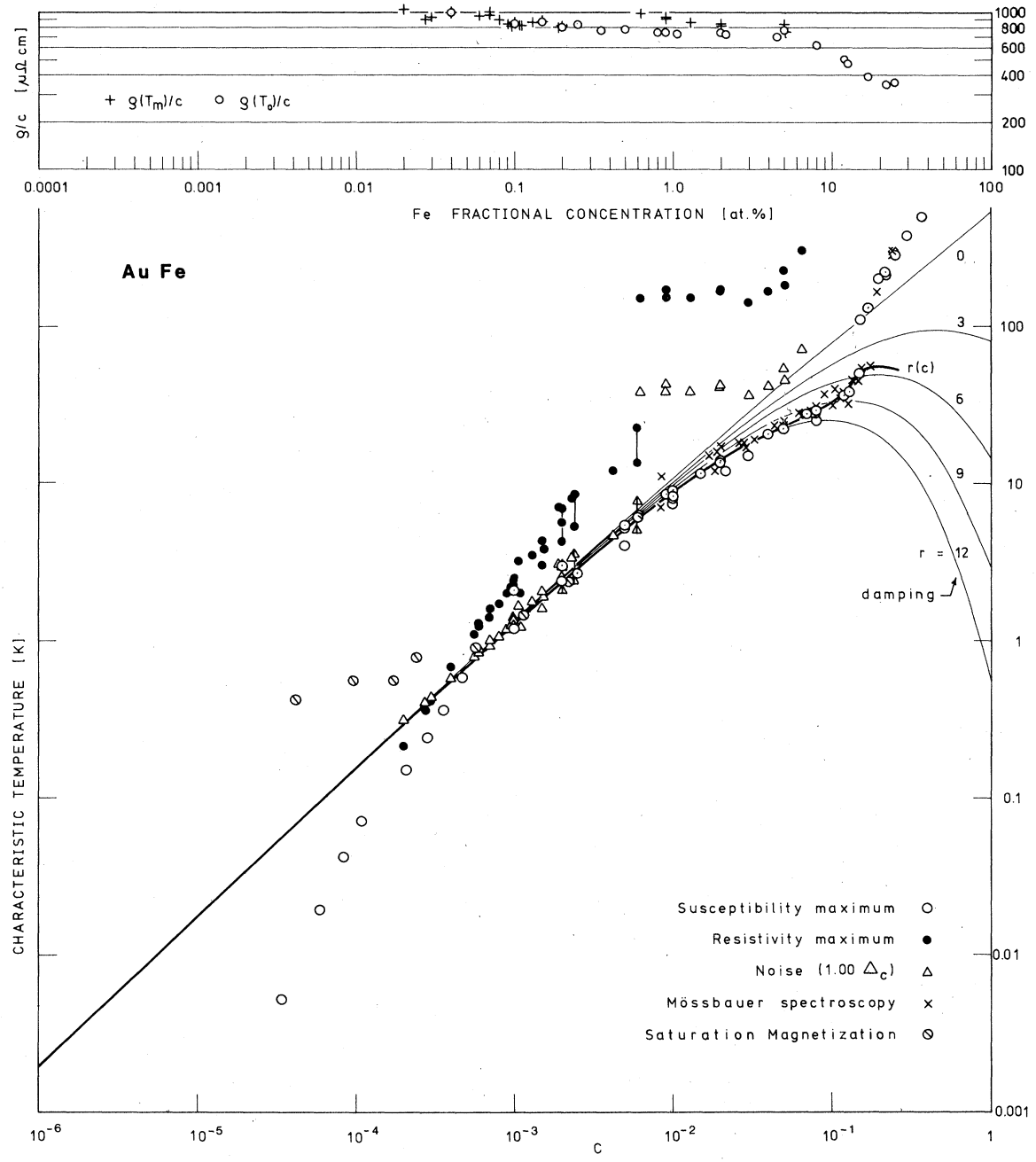

FIG. 1. Characteristic temperature in *Au*Fe as a function of the fractional concentration of Fe. Signatures corresponding to data listed in Tables I–V: T_0 (○) susceptibility maximum, $T_{\dot{0}}$ (⊙) susceptibility cusp, $T_{\ddot{0}}$ (×) Mössbauer hyperfine splitting, T_H (⊘) high-field magnetization according to Eqs. (1) and (2), T_m (●) resistivity maximum, and Δ_c (Δ) noise temperature according to Eqs. (4) and (5). Points connected by vertical bars correspond to quench condensed films in various stages of annealing, according to Table V. In the upper section of the figure are shown values of the total resistivities at T_0 and T_m, divided by c. Full curves are theoretical results calculated according to Eqs. (6) and (7) for constant values of the damping parameter r, as indicated in the figure. The heavy curve with a concentration-dependent $r(c)$, obtained from the resistivity data in the upper section, is a fit of this theory to the data. For further discussion see the text. See also Fig. 2.

This simple situation may be complicated by the phase shift of the oscillations in $\mathcal{J}(R_{ij})$ that accompany the damping. The phase shift will depend on the specific properties of the system between pairs of spins $\vec{S}_i$ and $\vec{S}_j$, and may not be the same for the next pair $\vec{S}_i$ and $\vec{S}_{j'}$ in the sum over j, even though $R_{ij} = R_{ij'}$, because the system is not uniform but random. This means that steps in the random walk of $\vec{H}_{(i)j}$ may occur due to spins at the distance R_{ij} with lengths ranging from zero up to the maximum possible $\mathcal{J}(R_{ij})$ at this range. Hence $W_{(i)j}$ has some other form than the simple δ function. Obviously its second moment will still be of the order $[\mathcal{J}(R_{ij})]^2$, and the freezing temperature will still have the form (11). Therefore, de-

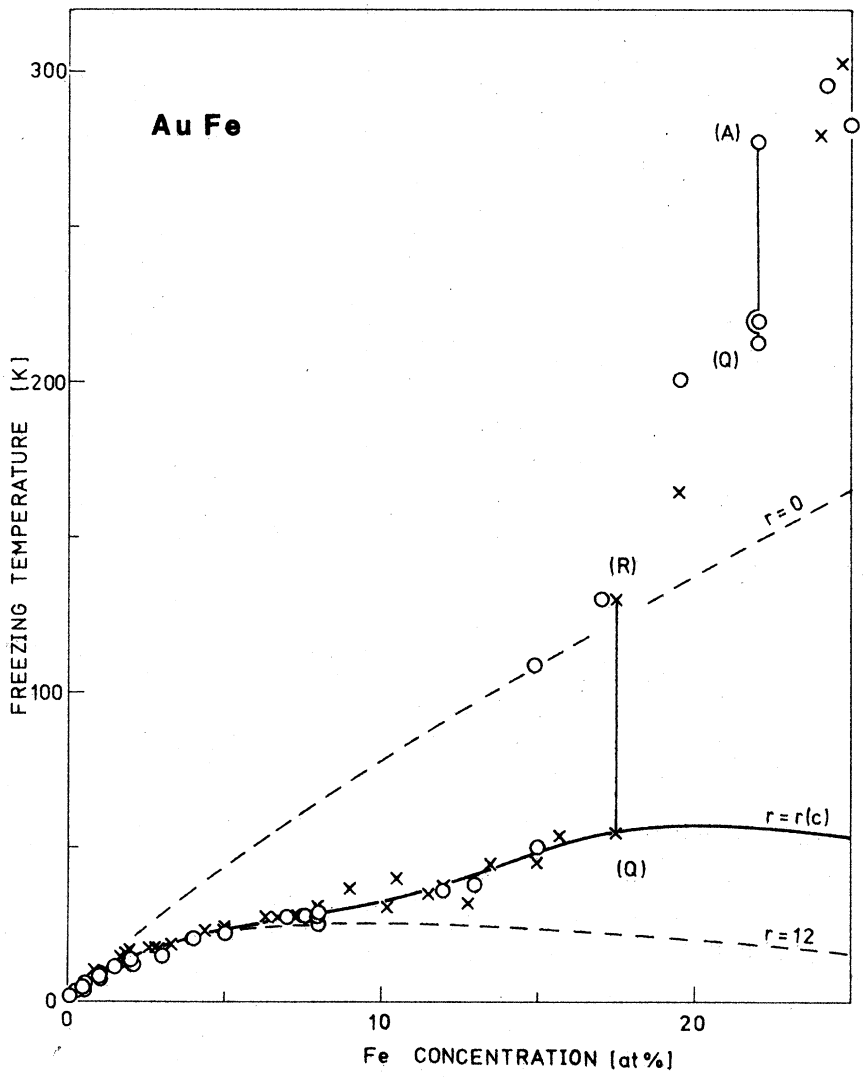

FIG. 2. Extract of Fig. 1 shown on linear concentration scale to emphasize the sharpness of the transition between the spin glass phase for $c \lesssim 17\%$ and the dilute ferromagnetic phase for $c \gtrsim 17\%$. The extreme sensitivity in the transitional region to the degree of compositional short-range order indicates a percolation threshold for short-range interactions. These effects of different metallurgical treatments are illustrated for 17.5% (Ref. 15) and 22% (Ref. 16), where Q = quenched, R = short-range ordered due to radiation enhanced diffusion, and A = aged for 1500 h at 24 °C. The heavy curve with the concentration dependent damping parameter $r(c)$ is the theoretical spin-glass freezing temperature incorporating the observed total resistivity (shown in top section of Fig. 1). Note the importance of the reduction in this resistivity, and hence in $r(c)$, in obtaining a good fit to the observed freezing temperatures between 5% and 17%. This is evidence for the dominance of the RKKY interaction in the freezing process persisting all the way up to the percolation threshold.

tails of $W_{(i)j}$ are of little importance for the freezing temperature.

To take an antagonist of the δ function, one may assume a Gaussian,

$$W_{(i)j}(\vec{H}_{(i)j}) = \frac{1}{\{2\pi\Delta(R_{ij})^2\}^{3/2}} e^{-|\vec{H}_{(i)j}|^2/2\Delta(R_{ij})^2} , \tag{12}$$

and take $\Delta(R_{ij})$ as one half times the envelope of $\mathcal{J}(R_{ij})$,

$$\Delta(R) = \frac{9\pi J^2(2I+1)^2}{2E_F(2k_FR)^3} e^{-R/\lambda} \tag{13}$$

in order to include virtually every $\vec{H}_{(i)j}$ within the fairly sharply defined maximum value of $\mathcal{J}(R_{ij})$ given by the envelope. This turns out to be a reasonable choice. It then follows[121] that $W_{(i)}$ is also a Gaussian,

$$W_{(i)}(\vec{H}_{(i)}) = \frac{1}{(2\pi\Delta_{(i)}^2)^{3/2}} e^{-|\vec{H}_{(i)}|^2/2\Delta_{(i)}^2} \tag{14}$$

and

$$\Delta_{(i)}^2 = \sum_{j\neq i} \Delta(R_{ij})^2 , \tag{15}$$

in accordance with the general result (A9) of Appendix A. Then again

$$(k_BT_0)^2 \propto \langle\Delta_{(i)}^2\rangle = \langle\sum_{j\neq i}\Delta(R_{ij})^2\rangle , \tag{16}$$

which is clearly not essentially different from (11). The average over spins (i), implicated by $\langle\ \rangle$, must be designed to take into account the concentration fluctuations in the local environment through a distribution of distances to the first neighboring spin[31] (for details cf. Ref. 7).

Computer simulations[122,123] of RKKY coupled spinglasses at zero temperature have shown very little directional correlation between spins even in the completely frozen ground state(s), with the exception perhaps of one or two neighbors of a given spin. In these calculations the possibility of random phase shifts due to mean-free-path effects were not considered, and they would be very difficult to estimate realistically, anyway. It therefore seems that the assumption of no directional correlations between pairs of spins is justified for the majority of pairs in the spin glass. Obviously, it ceases to hold if dilute ferromagnetic order occurs, and more massive forms of short-range order may also be beyond such a simple formulation. Comparison of the observed concentration dependence of the characteristic temperatures with the above simple calculation, that neglects directional correlations, should therefore make such effects stand out with clarity, when and if they actually occur. This is what happens and will be discussed in the concluding section.

At temperatures above the freezing the spins become uncorrelated and the field $\vec{H}_{(i)}$ fluctuates in time, as it may do to some extent at any finite temperature. A calculation of the spin flip transition rate, or the noise temperature Δ_c, in fact gives[31]

$$\Delta_c \propto \langle\sum_{j\neq i}\mathcal{J}(R_{ij})^2\rangle^{1/2} , \tag{17}$$

which is appropriate in the light of the above qualitative discussion, and directly explains the coincidence of Δ_c and T_0 observed in Fig. 1.

In the model of Edwards and Anderson[17] one *explicitly* disregards directional correlations between spins on different sites, also in the frozen spin glass state. This is instituted by the introduction of distributions of exchange couplings

$$P(\mathcal{J}(R_{ij})) = \frac{1}{[2\pi\Delta(R_{ij})^2]^{1/2}} e^{-\mathcal{J}(R_{ij})^2/2\Delta(R_{ij})^2} . \tag{18}$$

When this is compared with the three-dimensional distribution of fields (12), it becomes easy to appreciate that the freezing temperature is indeed given by

$$k_BT_0 = b_S\langle\sum_{j\neq i}\Delta(R_{ij})^2\rangle^{1/2} , \tag{19}$$

where b_S for quantum spins[18,124] was given in (7). Using (13) this expression gives the result (6).

If one restricts the model of Edwards and Anderson to a *single* Gaussian distribution of couplings instead of (18) there is a difficulty in applying it to the RKKY interaction, which has a distribution that diverges for small couplings, due to its infinite range and oscillations. This is, however, irrelevant with respect to the present calculation of T_0 due to the use of *infinitely many* Gaussians in (18), each pertaining only to a given distance R_{ij}. The argument is explained in more detail in Appendix B.

In (6) the damping parameter r is proportional to the *total* resistivity ρ, expressed in terms of the contribution per magnetic impurity $\bar{\rho} = \rho/c$. This is to be understood as the total measured resistivity at the relevant temperature, as described in the previous section. The magnetic impurity contribution, that is roughly proportional to c, is usually the dominant cause of resistance. In terms of the differential scattering cross section, $\sigma(\theta)$, the resistance should be roughly proportional to the total *transport* cross section

$$\sigma_{tr} = 2\pi\int_0^\pi d\theta\,(1-\cos\theta)\sin\theta\,\sigma(\theta) . \tag{20}$$

On the other hand, the relevant mean free path to enter the RKKY interaction in the damping exponent should rather be inversely proportional to the *ordinary* cross section

$$\sigma = 2\pi\int_0^\pi d\theta\sin\theta\,\sigma(\theta) . \tag{21}$$

Therefore, the damping parameter is given in terms of σ, while ρ is given by σ_{tr}, and the relation for a spherical Fermi surface is

$$r \equiv \frac{2a_0}{\lambda c}\left(\frac{3}{16\pi}\right)^{1/3} \simeq \frac{8}{a_0^2}\left(\frac{3}{16\pi}\right)^{1/3}\sigma = \frac{\sigma}{\sigma_{tr}}\frac{4}{a_0^2}\left(\frac{3}{2\pi}\right)^{1/3}\sigma_{tr} = \frac{\sigma}{\sigma_{tr}}\frac{4e^2\rho/c}{\hbar k_F a_0^2}\left(\frac{3}{2\pi}\right)^{1/3} = \frac{\sigma}{\sigma_{tr}}\frac{4e^2\bar{\rho}}{ha_0} \quad . \tag{22}$$

In expression (7) the correction factor β is therefore the ratio of cross-sections σ/σ_{tr}. As I cannot calculate it from first principles, β is kept open as a fitting parameter in the expression for $k_B T_0$. It only affects the overall strength of the damping, but not its concentration dependence which follows the measured $\bar{\rho}$ shown in Fig. 1.

The magnitude of β can be estimated expressing the cross-sections in terms of phase shifts in the partial wave expansion (in standard notation)

$$\sigma(\theta) = k_F^{-2}\left|\sum_{l=0}^{\infty}(2l+1)e^{i\eta_l}\sin\eta_l P_l(\cos\theta)\right|^2 \quad .$$

The standard results of the integrations (20) and (21) are

$$\sigma_{tr} = 4\pi k_F^{-2}\sum_{l=1}^{\infty} l\sin^2(\eta_{l-1}-\eta_l) \quad ,$$

$$\sigma = 4\pi k_F^{-2}\sum_{l=0}^{\infty}(2l+1)\sin^2\eta_l \quad .$$

Suppose only the first l_0 phase shifts are nonvanishing and all of the same magnitude, equal to η. One gets

$$\sigma_{tr} = 4\pi k_F^{-2}(l_0+1)\sin^2\eta \quad ,$$

$$\sigma = 4\pi k_F^{-2}\sin^2\eta(l_0+1)^2 \quad .$$

So in this somewhat artificial situation $\sigma/\sigma_{tr} = l_0+1$. One also easily shows that if only a single phase shift is active, one gets $\sigma_{tr} = \sigma$. It should be reasonable then to expect values of β bigger than unity and perhaps around 3 for $3d$ impurities. This agrees reasonably well with what is found in *Au*Fe and *Cu*Mn (see Sec. IV and Ref. 6).

IV. DISCUSSION

It has so far been unclear what is the precise upper limit of concentrations where the RKKY interaction dominates. Generally, but with the exception of the work mentioned in the Introduction, the upper limit is set about 1%. Beyond this the situation is expected to become complicated by the presence of clusters of spins coupled by additional direct exchange between nearest neighbors in increasing quantity. It is not entirely clear how this influences the freezing process, which is presently not well understood (in the sense of consensus) at any concentration.[2]

In Figs. 1 and 2 is shown a set of curves calculated from (6) for constant values of the damping parameter r. These curves would correspond to a concentration independent $\bar{\rho} = \rho/c$, according to (7). Since the total resistivity is then assumed proportional to c, this is a situation dominated by scattering by the spins themselves, i.e., *self*-damping.[30] The effect is most pronounced at high concentrations. Comparing with the osberved $\bar{\rho}(T_0)$ and $\bar{\rho}(T_m)$, shown in the top section of Fig. 1, proportionality is fairly well observed up to about 8%, where $\bar{\rho}(T_0)$ begins to drop rather rapidly, having decreased by a factor of 2 at 15%–20%. This drop in the resistance should therefore produce a similar drop in the damping parameter by a factor of 2, and hence cause a transition across the constant r curves in the upward direction between 8% and 15%–20%. This is precisely what happens with T_0 and $T_{\ddot{0}}$. Note that the observation can in fact be made before one applies any fitting procedure to find the two parameters of the theory, A and β. It is therefore *a plain correlation between different measurements, which immediately shows that the coupling between the spins is sensitive to the mean free path of the conduction electrons.* Therefore, it must be taking place via the RKKY interaction, as direct-exchange coupling does not involve the conduction electrons.

The parameter values were determined before[7] considering data for T_0 exclusively. No changes are necessary to incorporate the new data included here. A good fit is obtained with $A/k_B = 530$ K and $\beta = 4$ using the observed concentration dependence of $\bar{\rho}(T_0)$. The theoretical freezing temperature thus obtained is shown as the heavy curve in Figs. 1 and 2. At low concentrations $\bar{\rho}(T_0) \simeq 780\ \mu\Omega$ cm, and with $a_0 = 4.07$ Å this corresponds to $r \simeq 12$.

Using (7) with $S = 1$ and $E_F = 5.5$ eV one obtains from the found value of A/k_B that

$$|J| = 0.23 \text{ eV} \quad . \tag{23}$$

This may be compared with the value of J found from the Kondo temperature $T_K = (E_F/k_B)\times \exp[-1/n(E_F)|J|]$, where $n(E_F)$ is the density of states/(eV atom) of both spin directions. Using $n(E_F) = 0.316\ \text{eV}^{-1}$ and $T_K = 0.19$ K one obtains

$$J = -0.25 \text{ eV} \quad . \tag{24}$$

The agreement between the two values of J is strong evidence for the RKKY interaction. This is further confirmed by the determination of J from the pressure

dependence of the resistivity, where a value was obtained that is considered very certain,[22,23]

$$|J| = 0.24 \text{ eV} \quad . \tag{25}$$

Here it is important to point out that when one derives Δ_c from T_m and T_K using (4) not only the vertical position of the points in the log-log plot of Fig. 1 depends on the value of T_K, but, what is more important, also the *slope* depends significantly on T_K. Hence it is *only* the value of T_K used here that will produce (i) the coincidence of Δ_c with T_0 and T_H as observed experimentally, and (ii) the simultaneous coincidence with the theoretical curve, which was already laid out before the introduction of Δ_c into the picture, by comparison with T_0 at higher concentrations.[7] Note that in Fig. 1 this theoretical curve has a slope ~ 0.9 in the appropriate region with little influence of damping. There are therefore two alternative conclusions one may draw from this result: *Either*, as in *Au*Fe here, one may consider T_K determined by other means, and then the data of Δ_c provide evidence for the concentration fluctuation effect (relation (4) is already verified independently by the pressure studies[3,4]); *or*, and this will be the strategy with *Cu*Mn, one may adjust T_K until Δ_c has the right slope and thereby determine the value of T_K. In *Au*Fe this would have given a fourth independent determination of J, equal to the value in (24). The accuracy with this method is not as good as with the pressure method,[22,23] however.

The numerous possibilities for quantitative cross checks of the theories involved are emphasized here because of the rather unique position of the *Au*Fe system among spin-glasses. No other system presently provides all these opportunities. For example, *Cu*Mn has a Kondo temperature essentially outside the directly accessible temperature regime.

Proceeding with the other fitting parameter, $\beta = 4$ is reasonable in the light of the argument given in the previous section. A rather critical test of the correctness of the theory regarding the way damping is included, and of this value of β, can be obtained from the measurements on quench-condensed films.[27,28] Here the variation of the resistance maximum and T_m is determined as a function of the annealing temperature. Data for $c = 0.2\%$,[27] $c = 0.24\%$,[28] and $c = 0.6\%$[28] are given in Table V. As the films have very high resistivities, more than a factor of 10 above bulk, a considerable variation of T_m, and hence of the corresponding Δ_c, is observed. Using the present theory of the damping effects one can account for the change in T_m by a change in Δ_c,[28] caused by a change in the damping due to the change in $\rho(T_m)$, following the annealing. In the case of Ref. 28 there is no significant difference between $\rho(T_m)$ and $\rho(4.2\text{ K})$, and the latter is used. Calculating r from (22) with $\beta = 4$ and bulk $a_0 = 4.07$ Å, one obtains the values given in Table V.

These data are compared with the calculated decrease according to (6) in Fig. 3. The agreement is satisfactory. Even the upward curvature is reasonably well accounted for by the theory. If the shallow minimum apparent in the data of Korn[27] at the highest r is a real effect, which cannot be decided with the present data, it does not seem to be explainable without the introduction of additional effects, such as changes in a_0 and/or T_K, that may be significant in the most disordered films.

In an early investigation Béthoux *et al.*[127] and Souletie *et al.*[32,128] studied the effects of adding up to 6-at.% Ti to the Au matrix. The resulting drastic reduction of the mean free path, leading to values of r as large as about 1000, causes a variety of changes in the properties. Among these is a reduction of T_0 at $c = 0.1\%$ to about 30% and 40% at $r = 555$ and 958, respectively, of its value at $r = 0$. The overall agreement with the present theory remains satisfactory, T_0 at $r = 600$ being predicted at 38% of the $r = 0$ value.

It is of interest also to consider the initial decrease

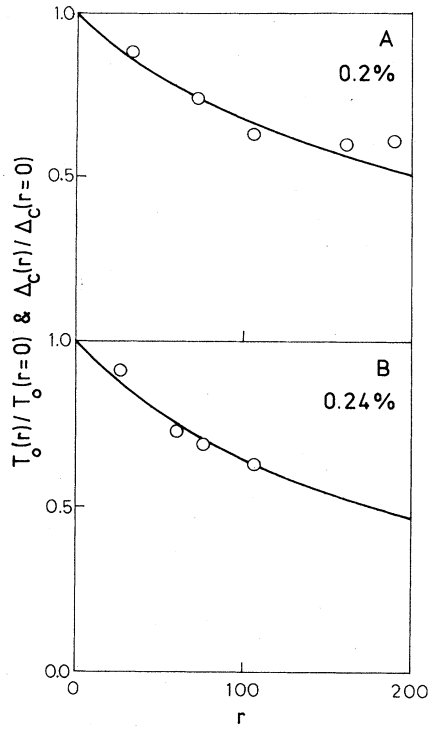

FIG. 3. Noise temperature Δ_c (○) corresponding to resistivity maxima in quench-condensed films (Table V). Annealing at different temperatures changes both Δ_c and the total resistivity of the film, hence the damping parameter r. Full curves are the theoretical dependences of T_0 or Δ_c on r. (A) Data of Korn (Ref. 27), $c = 0.2\%$, $\Delta_c(r=0) = 3.42$ K. (B) Data of Buchmann *et al.* (Ref. 28), $c = 0.24\%$, $\Delta_c(r=0) = 3.85$ K. For further details see the text.

for small r. From Appendix C one gets the theoretical result (C9) that should hold for Δ_c as well:

$$\frac{1}{T_0}\frac{\partial T_0}{\partial r}\bigg|_{r=0} \simeq \frac{3}{4}\frac{\Gamma(\frac{1}{3})c^{2/3}-3c}{\gamma+\ln(c)} \quad .$$

For $c=0.24\%$ this gives the value -5.6×10^{-3}, while the extrapolated initial slope of Δ_c from the data of Buchmann *et al.*[28] is -4.8×10^{-3}. The curvature for higher values of r evident in Fig. 3 indicates that the linear approximation is ceasing to be valid for $r \gtrsim 50$, where $rc \gtrsim 0.12$ is no longer $\ll 1$. The effect of the curvature is to make the data taken at finite r produce a somewhat smaller extrapolated initial slope than the true.

The agreement between the observed and calculated dependence on r propagates to the value $\beta=4$, since it enters via r as a constant of proportionality. The effects of damping on T_0 and Δ_c in quite different experiments are therefore consistently accounted for, both as regards the dependence on c for "constant" r and on r for constant c.

V. CONCLUSIONS

It has been found that the theoretical expression (6) gives a good quantitative description of the concentration and mean-free-path dependence of the characteristic temperatures over a considerable range of intermediate concentrations. One can therefore conclude that the RKKY interaction is a common source of the various physical properties of alloys of *Au*Fe throughout this composition range.

At the lowest and highest concentrations there are deviations from the theoretical expression. Even though one can have a fairly good idea about what the reasons for these deviations are, it is clear that further work, both experimental and theoretical, is needed before a quantitative understanding of even the simple kind described in this paper is obtained.

At *low concentrations* the calculation shown as the heavy curve in Fig. 1 predicts a nonlinear concentration dependence due to the concentration fluctuations. As emphasized above, values of Δ_c derived from resistance maxima fall exactly on the theoretical curve, but at the lowest concentrations this is no longer the case with T_0 and T_H. This is most likely due to the Kondo effect, as remarked in Sec. II. Evidence is that the deviations take place when $T_H \sim T_0 \sim T_K$ and that T_m is rapidly decreasing with decreasing c, and in fact cuts through Δ_c, in the same range of concentrations.[3] T_m is a function of both the RKKY interaction strength Δ_c and the Kondo temperature T_K ,

$$T_m = T_m(\Delta_c, T_K) \quad ,$$

and, as it happens in this case, for *all* concentrations. The conclusion is therefore that at *low* concentrations a similar complication takes place in the other characteristic temperatures

$$T_0 = T_0(\Delta_c, T_K) \quad ,$$

$$T_H = T_H(\Delta_c, T_K) \quad .$$

There exists no consistent quantitative theory of these effects. The above qualitative interpretation is supported by the analysis of[6] *Cu*Mn and other low-T_K systems, where such deviations do not occur until at much smaller concentrations and temperatures. *Cu*Mn has a T_K of the order of magnitude of 1 mK.

At *high concentrations* resistivity data for $\bar{\rho}(T_0)$ have been obtained up to 26%. After the decrease by a factor of 2 from 5% to 20% it seems to level off above 20% (not considered in Ref. 7), whereby the damping parameter settles about $r \simeq 6$. The coincidence of the corresponding theoretical curve with the observed T_0 and $T_{\ddot{0}}$ in Fig. 2, quantitatively supports Murani's suggestion of RKKY dominated spin-glass behavior extending to high concentrations. Furthermore, this comparison leads to the interesting conclusion that the observed values of T_0 and $T_{\ddot{0}}$ above 17% rather suddenly cease to be well described by the RKKY based calculation and increase steeply above the predicted values. A very similar observation has been made in *Cu*Mn.[6] This takes place at the nearest-neighbor percolation threshold[129] and may therefore in *Au*Fe be interpreted as the emergence of dilute ferromagnetism due to direct exchange coupling between nearest neighbors. This violates the assumptions behind the calculation described in Sec. III. Firstly, the direct exchange was not included, and secondly, there are now long-range correlations between the spins, violating the isotropic random-walk assumption. The deviation between observations and the theory is therefore to be expected.

It is interesting to speculate about the relative importance of the two agents referred to above. The direct exchange was not included at any concentration, and still despite the growing number of near-neighbor pairs it was found that the RKKY calculation remains quantitatively sufficient all the way up to the percolation threshold. This indicates that a nearest neighbor with an additional direct exchange coupling is not *in itself* a significant factor in comparison with the influence of the RKKY interactions with *all* the spins *everywhere* in the system. This somewhat unusual situation is a well-known consequence of the R^{-3} dependence of the RKKY interaction, which means that at increasing distances the decrease of interaction strength is exactly compensated by the growing number of spins present in a shell at any given range. This is the idea of the scaling laws in concentration.[29] It therefore seems that when suddenly the deviation occurs it is due to the second agent, namely, the occurrence of directional correlations.

One is therefore required to explain how, in the

presence of both the two interactions RKKY and direct exchange, with the continuous variation of the parameter c, at a critical concentration there is a changeover from the RKKY dominated spin glass to the dilute ferromagnet.

An interesting possibility of investigating the changeover region experimentally is to apply pressure in measurements of T_0 and $T_{\ddot{0}}$. In *Au*Fe it was found[4,23] that J varies linearly with the volume reduction $v=(V_0-V)/V_0$ due to pressure,

$$J(v)\simeq J(0)(1+\epsilon v) \ ,$$

with $J(0)=-0.24$ eV and $\epsilon=1.4$. A corresponding increase in Δ_c was observed,

$$\Delta_c(v)\simeq\Delta_c(0)(1+2\epsilon v)$$

with $\Delta_c(0)=1.8$ K, which corresponds to $\Delta_c\propto J^2$ as given by (7). The same effect should therefore be observable in T_0 in $T_{\ddot{0}}$. Since the magnitude of the resistivity does not vary enough with pressure to significantly change the damping parameter r, the magnitude of this pressure effect would remain constant throughout the range where the RKKY interaction dominates T_0. In *Au*Fe a pressure of 100 kbar produces $v=4.8\%$ giving a relative increase of $\sim 2\epsilon v=13.4\%$, that should be clearly detectable. It is conceivable that this should change as the percolation threshold is crossed with increasing concentration. The manner of change should provide insight into the nature of the changeover phenomenon.

Theoretically, an analogous problem of change-over in the model of Edwards and Anderson was discussed by Sherrington and Southern.[18] In the present context the percolation theory of Smith[130,131] seems attractive. There the significance of percolation for the freezing of the spins is the emergence of an infinite cluster of spins linked up by interactions that are larger than the thermal energy k_BT. One may say that spins in groups of any size, linked up in this way, are "communicating," which does not necessarily imply that they are frozen into fixed directions. In the dilute systems this communication can only take place via the long-ranged RKKY interaction. With decreasing temperature the groups of communicating spins grow until eventually at the freezing temperature they percolate and form an infinite cluster.

With increasing concentration the number of nearest neighbors that couple via additional direct exchange increases. Hence, even though such spins may become communicating, *this* coupling alone cannot lead to the formation of any *infinite* cluster as long as the concentration is below the threshold for nearest-neighbor percolation, c_0. Therefore, even in this case freezing takes place only when possible by the RKKY couplings that eventually link up the little groups. Therefore, T_0 remains controlled by the RKKY interaction, as evidenced by the dependence of T_0 on the electron mean free path, at least until the nearest-neighbor threshold is reached. At these concentrations there may be a sort of competition between states of order with and without directional correlations. One may see in the results of Borg and Dienes[15] and of Maartense and Williams[16] (Fig. 2), which show an extreme sensitivity to compositional short-range order, some hint at such a state of affairs. In general it seems that dilute ferromagnetism in *Au*Fe rapidly becomes favorable with increasing $c-c_0$ and takes over rather suddenly at $\gtrsim c_0$.[132]

But it should be emphasized that the situation in the real systems, like *Au*Fe and *Cu*Mn, of having both ordinary direct exchange and RKKY indirect exchange is new and challenging. The phenomena associated with the changeover between the two states of order,[8-16,122] spin glass and dilute ferromagnetism, are not likely to be understood on the basis of theories that have only included one or the other of the interactions.

ACKNOWLEDGMENTS

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APPENDIX A

In order to calculate the second moment Ω of the probability distribution $W(\vec{H})$ of the total fields $\vec{H}=\sum_{j=1}^{N}\vec{H}_j$ that are the outcomes of random walks of N fields $\vec{H}_j$ consider the general formulation

$$W(\vec{H})=\int_{-\infty}^{\infty}\frac{d\vec{h}}{(2\pi)^3}e^{-i\vec{h}\cdot\vec{H}}A_N(\vec{h}) \ , \tag{A1}$$

$$A_N(\vec{h})=\prod_{j=1}^{N}\int_{-\infty}^{\infty}d\vec{H}_j\,e^{i\vec{h}\cdot\vec{H}_j}W_j(\vec{H}_j) \tag{A2}$$

given by Chandrasekhar.[121] $W(\vec{H})\,d\vec{H}$ is the probability of finding $\vec{H}$ in $(\vec{H}\,|\,\vec{H}+d\vec{H})$, and similarly $W_j(\vec{H}_j)\,d\vec{H}_j$ is the probability that the jth step of the field is in $(\vec{H}_j|\,\vec{H}_j+d\vec{H}_j)$. Note that each W_j may be different from the others. The second moment may be obtained from the inverse transform of (A1),

$$A_N(\vec{h})=\int_{-\infty}^{\infty}d\vec{H}\,e^{i\vec{h}\cdot\vec{H}}W(\vec{H}) \tag{A3}$$

by application of the Laplacian operator

$$\Delta_h=\frac{\partial^2}{\partial h_x^2}+\frac{\partial^2}{\partial h_y^2}+\frac{\partial^2}{\partial h_z^2} \ , \tag{A4}$$

whereby one easily shows that for $H^2=\vec{H}\cdot\vec{H}$

$$\Omega\equiv\int_{-\infty}^{\infty}d\vec{H}\,W(\vec{H})H^2=-\lim_{\vec{h}\to\vec{0}}[\Delta_hA_N(\vec{h})] \ . \tag{A5}$$

Now assume that each step takes place in a random

direction. Then W_j is isotropic in H_j space and is a function of only the length H_j. W_j must be normalized and its second moment must exist,

$$\omega_j \equiv \int_{-\infty}^{\infty} d\vec{H}_j \, W_j(\vec{H}_j) H_j^2 \quad . \tag{A6}$$

Then the gradient of the transform of W_j vanishes in the limit $\vec{h} \to \vec{0}$,

$$\lim_{\vec{h}\to\vec{0}} \vec{\nabla}_h \int_{-\infty}^{\infty} d\vec{H}_j \, e^{i\vec{h}\cdot\vec{H}_j} W_j(\vec{H}_j) = \int_{-\infty}^{\infty} d\vec{H}_j \, (i\vec{H}_j) \, W_j(\vec{H}_j) = \vec{0} \quad . \tag{A7}$$

This means that only the following terms contribute to (A5):

$$\lim_{\vec{h}\to\vec{0}} \sum_{j=1}^{N} A_N(\vec{h}) \frac{\int_{-\infty}^{\infty} d\vec{H}_j \, H_j^2 e^{i\vec{h}\cdot\vec{H}_j} W_j(\vec{H}_j)}{\int_{-\infty}^{\infty} d\vec{H}_j e^{i\vec{h}\cdot\vec{H}_j} W_j(\vec{H}_j)} \quad . \tag{A8}$$

Using normalization and (A6) this gives

$$\Omega = \sum_{j=1}^{N} \omega_j \quad . \tag{A9}$$

This relation is completely general. If, for example, the length of each step H_j was given with certainty, say,

$$H_j = \mathcal{J}(R_{ij}) \quad , \tag{A10}$$

then one could take

$$W_j(\vec{H}_j) = \delta(H_j - \mathcal{J}(R_{ij}))/4\pi[\mathcal{J}(R_{ij})]^2 \quad , \tag{A11}$$

whereby

$$\omega_j = [\mathcal{J}(R_{ij})]^2 \tag{A12}$$

and

$$\Omega = \sum_{j=1}^{N} [\mathcal{J}(R_{ij})]^2 \quad . \tag{A13}$$

Also, the relation (15) between the widths of the Gaussians (12) and (14) in the text is a special case of (A9), for which the second moments are related to the widths according to $\Omega = 3\Delta_{(i)}^2$ and $\omega_j = 3[\Delta(R_{ij})]^2$.

APPENDIX B

In an RKKY system where spins are randomly distributed on the lattice sites with exchange couplings

$$\mathcal{J}_{ij} = \mathcal{J}(R_{ij}) = \mathcal{J}_0 \left[\cos(2k_F R_{ij}) - \frac{\sin(2k_F R_{ij})}{2k_F R_{ij}} \right] (2k_F R_{ij})^{-3} \tag{B1}$$

between spins at sites (i) and (j) at the distance R_{ij}, the distribution of couplings has been shown to have a divergence for $\mathcal{J}_{ij} \to 0$, due to the infinite range and the oscillations.[125,126] It is therefore not obvious that a calculation of the physical properties using a single Gaussian distribution

$$P(\mathcal{J}_{ij}) = [1/\mathcal{J}_0(2\pi)^{1/2}] e^{-\mathcal{J}_{ij}^2/2\mathcal{J}_0^2} \quad , \tag{B2}$$

that does not contain this divergence, will lead to correct results.

In the calculation that is used in the present work[7] this difficulty has been avoided by the use of *infinitely many* Gaussians, namely, one for each range interval R_{ij} to $R_{ij} + dR_{ij}$,

$$P(\mathcal{J}_{ij}, \Delta(R_{ij})) = \frac{1}{\Delta(R_{ij})(2\pi)^{1/2}} e^{-\mathcal{J}_{ij}^2/2\Delta(R_{ij})^2} \quad . \tag{B3}$$

The use of Gaussian distributions is much better justified in this case, as for the finite-range interval there are certainly never infinitely many couplings of any strength, like zero. The exponential damping factor one could include with $\mathcal{J}_{ij}$ would modify the $\mathcal{J}_{ij} \to 0$ divergence, but is not a satisfactory solution of the problem. Using the Edwards-Anderson result for the freezing temperature then involves the infinite number of Gaussian widths, such that

$$k_B T_0 \propto \langle \sum_{j \neq i} \Delta(R_{ij})^2 \rangle^{1/2} \tag{B4}$$

involves a summation over the entire system of spins throughout the crystal. The width is identified with the envelope of the RKKY interaction as described in the text.

In order to show that the $\mathcal{J}_{ij} \to 0$ divergence is indeed taken into account in this method, one can calculate the *total* distribution by summing up the Gaussians (B3) over the entire volume. Converting to a volume integral starting at a shortest range a_0 of the order of the lattice constant and chosen for convenience such that $2k_F a_0 = 1$, one has

$$P(\mathcal{J}) = 4\pi \int_{a_0}^{\infty} dR \frac{R^2 P(\mathcal{J}, \Delta(R))}{4\pi a_0^3/3} \quad ,$$

with

$$\Delta(R) = \mathcal{J}_0/(2k_F R)^3 = \mathcal{J}_0(a_0/R)^3 \quad .$$

The integral is elementary and leads to

$$P(\mathcal{J}) = \left(\frac{\mathcal{J}_0}{\mathcal{J}} \right)^2 \frac{1}{\mathcal{J}_0(2\pi)^{1/2}} e^{-(\mathcal{J}/\mathcal{J}_0)^2/2} \quad ,$$

which is an ordinary Gaussian multiplied by the diverging factor.

APPENDIX C

The freezing temperature calculated in Ref. 7 and given by (6) and (7),

$$k_B T_0 = Ac \left[\frac{3}{c} \int_1^{\infty} \frac{dx}{x^4} e^{-rcx} [1 - e^{c'(1-x^3)}] \right]^{1/2} \quad , \tag{C1}$$

is a decreasing function with increasing damping parameter r for fixed concentration c. The initial rela-

tive decrease can be obtained in terms of integrals

$$I_n \equiv \int_1^\infty dx\, x^{-n}(1-e^{c'(1-x^3)}) \tag{C2}$$

for $n=3,4$ as follows;

$$\frac{1}{T_0}\frac{\partial T_0}{\partial r}\bigg|_{r=0} = -\frac{cI_3}{2I_4} \tag{C3}$$

by direct differentiation of (C1). Integration of (C2) gives for $n>1$

$$I_n = \frac{1}{n-1} - \frac{1}{3}(c')^{(n-1)/3}e^{c'}\Gamma\left(\frac{1-n}{3},c'\right) , \tag{C4}$$

where Γ is the incomplete gamma function. For $n=4$ one obtains

$$I_4 = \tfrac{1}{3}c'e^{c'}E_1(c') , \tag{C5}$$

where E_1 is the exponential integral. For $n=3$ one may use the expansion

$$\Gamma(a,x) = \Gamma(a)\left[1 - x^a e^{-x}\sum_{n=0}^{\infty}\frac{x^n}{\Gamma(a+n+1)}\right] \tag{C6}$$

to obtain

$$I_3 = \tfrac{1}{2}\Gamma(\tfrac{1}{3})\left[(c')^{2/3}e^{c'} - \sum_{m=1}^{\infty}\frac{(c')^m}{\Gamma(m+\frac{1}{3})}\right] . \tag{C7}$$

Then for $c'=c/(1-c)$ Eqs. (C5) and (C7) inserted in (C3) gives the decrease of T_0 with r for a fixed c. One should caution against indiscriminate use of this expression for arbitrary values of r and c, because the integral in (C1) is not an analytic function of r and c. The nature of the nonanalyticity can be seen by considering the first term separately,

$$\int_1^\infty dx\, x^{-4}e^{-rcx} \doteq E_4(rc)$$
$$= \tfrac{1}{6}(rc)^3[\ln(rc) - \psi(4)]$$
$$- \sum_{\substack{m=0\\ \neq 3}}\frac{(-rc)^m}{(m-3)m!} .$$

The leading terms dominate and give an initial linear decrease with slope (C3) provided that

$$rc \ll 1 . \tag{C8}$$

As r can be considerably larger than unity this condition is not necessarily satisfied at any c. In such cases the dependence on r acquires curvature, and one must resort to numerical integration.

The leading contributions when $c\to 0$ to I_3 and I_4 are given by

$$I_3 \simeq \tfrac{1}{2}\Gamma(\tfrac{1}{3})c^{2/3} - \tfrac{3}{2}c + O(c^{5/2})$$

and

$$I_4 \simeq -\tfrac{1}{3}c[\gamma + \ln(c)] + O(c^2) ,$$

where $\Gamma(\frac{1}{3}) = 2.67894$ and $\gamma = 0.57722$. Thus one obtains for $c\to 0$

$$\frac{1}{T_0}\frac{\partial T_0}{\partial r}\bigg|_{r=0} \simeq \frac{3}{4}\frac{\Gamma(\frac{1}{3})c^{2/3} - 3c}{\gamma + \ln(c)} . \tag{C9}$$

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