Mean-free-path effects in the resistivity of *Au*Fe thin-film spin-glasses

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The low-temperature resistivity of *Au*Fe thin-film spin-glasses ($c = 0.24$-, 0.6-, 6.0-at.% Fe) is investigated after quench condensation in ultrahigh vacuum, and after annealing at different temperatures. By this method the mean free path (mfp) λ of the conduction electrons is varied, and the influence of the variation of λ on the low-temperature resistivity anomaly can be studied. The shortening of the mfp results in a damping of the Ruderman-Kittel-Kasuya-Yosida interaction strength, expressed by the de Gennes factor $\exp(-r/\lambda)$. With decreasing mfp the resisitivity maximum at T'_{max} is shifted to lower temperatures. A comparison with bulk-material data and a discussion of the dependence $T_{max}(\lambda)$ within the framework of different theoretical models allow predictions about the variations of the spin-glass freezing temperature T_0 with the mfp. Changes between 10% and 100% are deduced, depending on the impurity concentration and the chosen model. Moreover, the λ dependence of the slope of $\Delta\rho(T)$ in the linear-T region below T_{max} is discussed. The absolute value of the coefficient A of the spin-glass typical $T^{3/2}$ region and its observed variation with the mfp can be explained by the theory of Rivier and Adkins, which also predicts our observation of increasing slope and extension of a T^2 region in the resistivity at very low temperatures with decreasing mfp. The occurrence of a minimum in $\Delta\rho(T)$ as found in films with very short mfp at temperatures below the T^2 range is believed to be due to residual magnetism of single impurities not participating in the freezing process.

I. INTRODUCTION

The properties of the so-called spin-glasses, dilute alloys containing magnetic impurities in concentrations above the Kondo regime, are currently a subject of considerable interest.[1-4] Especially the question of whether there exists a phase transition in a spin-glass at a characteristic temperature T_0 is still unsolved. So far, from an experimental point of view, there is a sharp transition temperature (cusp) present only in the zero-field ac susceptibility, although measurements of the Mössbauer effect and μ^+ precession possibly yield the existence of a well-defined T_0, too. On the other hand, measurements of the specific heat, resistivity, and thermopower show no sharp transition in the temperature region around T_0. These experiments therefore give more support to the non-phase-transition models, which explain the changes in the properties of a spin-glass as a freezing process of magnetic clusters.[5,6] The magnetic moments of the clusters—originating from the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions between the magnetic impurities—are free to rotate at temperatures above T_0, whereas below T_0 they freeze progressively, depending on their size, into random directions. Therefore the freezing does not result in an equilibrium state, but a spin-glass appears to be metastable, e.g., to the application of an external magnetic field.[7,8]

As mentioned above the freezing temperature T_0 cannot be deduced directly from measurements of the resistivity. At high temperatures the incremental resistivity first increases due to spin-flip scattering until it reaches a maximum.[9] The maximum temperature T_{max} roughly marks the temperature range in which the clusters start growing. Due to the progressive freezing out of spin-flip scattering the resistivity decreases below T_{max} following different power laws in T. The occurrence of a maximum in $\Delta\rho(T)$ is thus the result of two competing physical mechanisms: the Kondo effect and the RKKY interaction.

Since the RKKY interaction is mediated by the conduction electrons the strength Δ_{RKKY} of this interaction depends on the mean free path (mfp) of the conduction electrons. This was first pointed out by de Gennes,[10] who showed that a shortening of the mfp damps the RKKY interaction. The damping of the RKKY amplitude can mainly be expressed by a factor $e^{-r/\lambda}$, where r is the distance from a magnetic atom, and λ is the mfp. Consequently, the properties of a spin-glass depend on the mfp. The aim of the present work is to study this influence experimentally.

We think the best method to vary the mfp in alloys is to evaporate the materials at low temperatures. Due to the high degree of lattice disorder introduced by the quench condensation, the mfp is very short.

Afterwards it can be increased by annealing of the sample until the impurity limited bulk value (self-damping) is reached.[11,12] For our investigations we have chosen the *Au*Fe system, because it is the most widely studied spin-glass system and a large number of data is available for the comparison with the films.

The present study is mainly concerned with the question of how the low-temperature resistivity is influenced by changes of the excitations of the frozen spins with the mfp. We will show that when λ is short there exists a T^2 dependence in the resistivity at $T \ll T_{max}$. This behavior has been predicted theoretically by Rivier and Adkins.[13] These authors also predict that the T^2 law is replaced by a $T^{3/2}$ dependence when the mfp is increased. This change is demonstrated experimentally in the present work, and makes questionable earlier statements asserting that the occurrence of the $T^{3/2}$ law in the resitivity is really a typical spin-glass feature. Our results obviously support the idea that the $T^{3/2}$ law is only a transition between a T^2 region at low temperatures (reminding of localized spin fluctuations) and the resistivity maximum. A T^2 dependence on dilute *Au*Fe bulk alloys has been found recently by Laborde and Radhakrishna.[14]

Further on, we will study the changes of the coefficients of the different power laws in the incremental resistivity $\Delta\rho(T)$ as a function of the mfp. Since a comprehensive theoretical description of the behavior of $\Delta\rho(T)$ of a spin-glass so far does not exist, we will discuss these results separately within the light of different theories.

Finally, we will analyze in detail the changes of the freezing temperature T_0 with the mfp $T_0(\lambda)$. With respect to the question whether there is a phase transition or glass transition it is interesting to note that in a spin-glass T_0 does not change when a magnetic field is applied (cusp in $\chi(T)$ rounds off), nor does it depend on the measuring time.[7,8] For the latter reason Smith[15] considers T_0 to be a "percolation temperature." With decreasing temperature the average cluster size grows and the susceptibility cusp is due to the null response of the infinite cluster to an applied field when the percolation limit is reached. T_0 therefore depends mainly on the impurity concentration and the RKKY interaction strength Δ_{RKKY}, which itself is damped by a shortening of the mfp.

Although one cannot study the dependence $T_0(\lambda)$ directly by resistivity measurements, an investigation of the variation $T_{max}(\lambda)$ allows a determination of the changes of T_0 with λ by applying a theory of Larsen.[16] Previously we have discussed[17] the shift of T_{max} with the mfp for a low concentrated *Au*Fe film within the framework of this theory. In the meantime new theoretical work has become available. We shall discuss the former results and new data on higher concentrated films in detail and compare them to recent theoretical models by Larsen[18] and by Kinzel and Fischer.[19–21] For example, numerical calculations of $T_0(\lambda)$ can be done by using the T_0 values from the bulk susceptibility measurements and the experimentally determined values of λ as parameters.

II. EXPERIMENTAL

The experiments have been carried out in an ultrahigh-vacuum ^{3}He cryostat, earlier described in detail.[22] A highly disordered alloy film is prepared by successive flash evaporation of small pellets of the thin rolled alloy material onto a helium cooled quartz substrate. About 200 pellets have to be evaporated to condense a film of roughly 1000 Å in thickness. During the evaporation process the vacuum remains better than $\sim 5 \times 10^{-8}$ Torr. In order to study the influence of changes in the mfp of the conduction electrons on the low-temperature anomalies, the film is stepwise annealed at different temperatures and recooled for measurement.

The resistance is measured using a standard four-probe dc technique. The setup has such a high stability that the relative accuracy of the measurement is greater than 2.5×10^{-5} during a period of 3 hours. This is the time necessary for measuring the low-temperature resistance after each annealing step. Details have been described earlier.[23] For a comparison of different films with each other and with appropriate bulk alloys, one has to know the resistivity of the films. Therefore, it is necessary to determine the geometry factor, and thus especially the film thickness, as accurately as possible. Using a mask technique, the films are given a meander geometry, resulting in an overall 110-mm length and 0.3-mm width. These dimensions can be determined with relatively high accuracy. Exact values for the film thickness are more difficult to achieve. Within the present work, the film thickness is determined optically by a multiple beam interference method. For alloy films with high impurity concentrations this method gives more satisfying results than calculating the thickness from the temperature-dependent part of the resistance, as discussed by von Bassewitz and von Minnigerode.[24] The discrepancies between the results of the two methods are mainly due to deviations from Matthiessen's rule (DMR).

The magnetic impurity concentration is determined from the residual resistivity ratio for the bulk alloy $\Delta r = 7.4\ \mu\Omega$ cm/at.%. For the films we take the difference between the residual resistivities of a room-temperature-annealed *Au*Fe film and a respective pure Au layer, and compare it to the above value. The concentration values found in this way correspond within ±2% to the nominal concentration.

Most important for the analysis of the temperature dependence of the resistivity as measured in our disordered films and its quantitative comparison to

bulk material values is the knowledge of the correct impurity contribution $\Delta\rho(T)$. For bulk samples this contribution is normally obtained by subtracting the pure matrix part:

$$\Delta\rho_b(T) = \rho_b(T)_{\text{alloy}} - \rho_b(T)_{\text{matrix}}$$

It should, however, be mentioned that even for bulk alloys this method is not correct, because it does not include the DMR. Therefore, one should write: $\Delta\rho_b(T) = \rho_b(T)_{\text{alloy}} - \rho_b(T)_{\text{matrix}} - \rho_b^{\text{DMR}}(T, \rho_0)$, since $\rho_b^{\text{DMR}}(T, \rho_0)$ is also a function of the residual resistivity ρ_0.[25]

For the disordered films this dependence of the matrix part on ρ_0 is of great importance, since ρ_0 changes drastically with the degree of disorder. The latter is difficult to control in detail; two different films having the same ρ_0 can hardly be produced separately. Therefore, we take the following procedure to overcome these difficulties. We define

$$\Delta\rho(T) \equiv \rho(T)_{\text{alloy}} - \rho_{\text{Au}}(T, \rho_0) \ ,$$

where $\rho_{\text{Au}}(T, \rho_0)$ is the temperature-dependent part of the resistivity of the disordered *pure* Au film with the same ρ_0 as the alloy film.

For the determination of $\Delta\rho(T)$ the procedure is as follows. First, we measure the resistivity of a pure Au film after quench condensation and after different annealing steps, i.e., we determine $\rho_{\text{Au}}(T, \rho_0)$ for different ρ_0 values. A double logarithmic plot of $\rho_{\text{Au}}(T, \rho_0)$ vs ρ_0 shows that for each fixed temperature T_x the $\rho_{\text{Au}}(T_x, \rho_0)$ data can be represented by straight lines. This means we have received a set of straight lines giving ρ_{Au} as a function of ρ_0 with T_x as the parameter. Second, we measure the resistivity of the alloy films, again with different ρ_0. Since the residual resistivities of the alloy films do not coincide with those of the pure films, we determine the respective $\rho_{\text{Au}}(T_x, \rho_0)$ from the diagram discussed above. An advantage of this method is that the DMR in the alloy films are subtracted to a very far extent.

Finally we turn to the determination of the electronic mfp λ. The λ values for the films and the bulk samples given in this paper have been determined from the resistivity at 4 K using the relation (free-electron gas)[26]:

$$\rho\lambda = mv_F/ne^2 = 8.36 \times 10^{-12} \ \Omega\,\text{cm}^2 \ .$$

For the Au + 0.24-at.%-Fe alloy we find a mfp of 460 Å for the bulk alloy. By quench condensation this value is reduced to 50 Å for the film sample after the first annealing step. For the Au + 6-at.%-Fe alloy the mfp for the bulk sample is only some 20 Å. For this concentration the mfp can be changed only by about 30% by quench condensation. Thus it is obvious that true mfp effects can be studied best in low concentrated magnetic alloy films, as a wide range in the mfp is accessible.

III. RESULTS AND DISCUSSION

A. General behavior of the resitivity and investigation of the region $T > T_{\text{max}}$

Figure 1 shows a set of curves of the incremental resistivity $\Delta\rho$ as a function of temperature for a Au + 0.24-at.%-Fe alloy film with λ as parameter, together with the data for the corresponding bulk samples.[27] An interesting feature concerning the appropriate subtraction of the pure Au resistivity can be seen from this graph. If for the bulk sample the temperature-dependent part for pure Au as given by Mydosh *et al.*[28] is subtracted, there still is a minimum present in the $\Delta\rho(T)$ curve for $T > T_{\text{max}}$, which is due to DMR. To account for the phonon scattering and DMR, Schilling *et al.*[29] proposed to subtract from the resistivity measured an amount equal to $2.2\rho_{\text{phonon}}$. For the bulk alloy this results in the crosses shown in Fig. 1. Around T_{max} these values coincide with those obtained by subtracting the contribution $\rho_{\text{Au}}(T, \rho_0)$, as determined for the corresponding ρ_0 from our pure Au film (open circles in Fig. 1). This demonstrates that the method to determine the incremental contribution $\Delta\rho$ as discussed in Sec. II accounts to a great extent for the subtraction of the DMR.

As already mentioned in the introduction, the exponential factor $e^{-r/\lambda}$ damps the RKKY amplitude and therefore, with decreasing mfp λ, the impurity-impurity interactions are progressively reduced. Besides the influence of this damping on the spin-glass properties of the *Au*Fe system, which will be discussed below, changes of the mfp also alter the resistivity in the temperature regime $T > T_{\text{max}}$, where the Kondo effect causes the increase of the resistivity with falling temperature. As can be seen in Fig. 1 for $T > T_{\text{max}}$ the maximum slope of $\Delta\rho(T)$ increases from 0.28 $n\,\Omega$ cm/K for the bulk alloy to 1.4 $n\,\Omega$ cm/K for the film with $\lambda = 50$ Å. This can be qualitatively understood in the following way. The damping of the RKKY amplitude by the reduction of λ results in the onset of the impurity interactions at progressively lower temperatures. Thus, the spin scattering is higher, leading to an increase of the slope in the region $T > T_{\text{max}}$ and to a shift of T_{max} to lower values. Finally even the "one-impurity limit" is reached in our relatively high concentrated *Au*Fe film when λ is short. This is demonstrated in Fig. 1, where the solid curve gives the "one-impurity limit" for the *Au*Fe system, as calculated by Laborde[30] using the model of Souletie.[31] For the film with $\lambda = 50$ Å, this one-impurity limit is approached for temperatures $T \geq 15$ K.

Quantitatively, however, a detailed analysis of the observed variation in the slope is rather difficult, because we actually deal with four competing contributions: (i) the RKKY damping due to the reduction of λ (this enhances the slope for $T > T_{\text{max}}$ as just discussed); (ii) the DMR, being important in this tem-

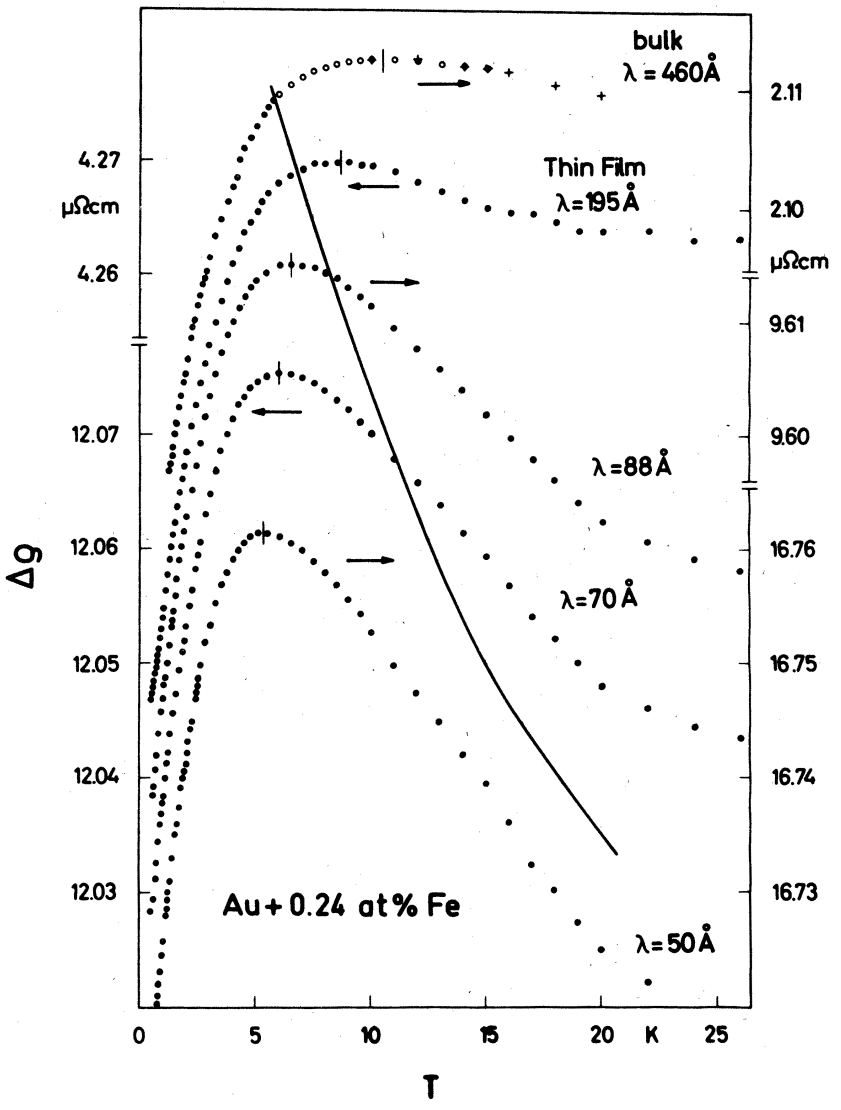

FIG. 1. Resistivity $\Delta\rho(T) = \rho_{\text{alloy}} - \rho_{\text{matrix}}$ vs temperature T for the alloy Au + 0.24-at.% Fe. The upper curve shows the resistivity of the bulk sample as measured by P. J. Ford (Ref. 27). (For explanation of the different symbols see text.) The four lower curves show the resistivity for the film after different annealing steps. The mfp values for the different curves are given in the graph. The position of the maximum has been marked by vertical bars. The solid curve gives the "one impurity limit" for *Au*Fe as calculated by Laborde (Ref. 30) using the model of Souletie (Ref. 31).

perature range, but quantitatively difficult to estimate; (iii) the reduction of the Kondo temperature with increasing degree of disorder in thin films, as recently discussed by us for *Cu*Fe.[12] This would reduce the slope, although the effect is believed to be small, since for *Au*Fe T_K is small, and in a temperature range $T >> T_K$ changes in T_K with λ are of minor influence. (iv) According to a model proposed by Daybell and Yeo,[32] recently discussed in detail for *Zn*Mn alloys,[33] a concentration-dependent number of impurity pairs is formed at high temperatures, which do not contribute to the temperature-dependent resistivity. Certainly, when the RKKY interaction is reduced, the number of pairs at a given temperature is reduced, and this would lead to an increase of the slope in the discussed temperature range in the disordered film. However, in *Zn*Mn (and all the other magnetic bulk systems) the changes of the slopes in $\Delta\rho(T)$ for $T > T_{\max}$ with increasing impurity concentration (decreasing λ) are small compared to our *Au*Fe film.

B. Resistivity maximum and spin-glass freezing temperature

Probably the most striking change in the resistivity curves of Fig. 1 is the shift of the maximum temperature T_{max} to lower values when the mfp is reduced. The T_{max} values—marked by vertical bars in Fig. 1—together with those of the 0.6% alloy are listed in Table I. Maxima cannot be observed in the 6% sample, even after quench condensation when λ is small.

As already mentioned, the occurrence of a maximum in the resistivity as well as the shift of its position with changes in the mfp are qualitatively understood. The maximum originates from the competition of two different mechanisms; the resistivity increase due to the Kondo effect and the freezing out of spin-flip processes due to RKKY interactions. A damping of the RKKY interaction by the de Gennes factor reduces the coupling and thus T_{max}.

In a previous paper[17] we have analyzed the shift in T_{max} quantitatively within the framework of a theory by Larsen[16] (hereafter referred to as L I). This theory takes into account the Kondo effect (calculated to infinite order) and compares it to the RKKY interaction strength, expressed by a rms interaction parameter Δ_c. T_{max} is thus a function of Δ_c and T_K. An analytic expression (Eq. 79 in L I) allows a numerical calculation of the interaction parameter Δ_c for given T_{max} and T_K. It has been further argued by Larsen *et al.*[34] that an expression similar to the one for T_{max} should hold for the spin-glass freezing temperature T_0, i.e., $T_0 = T_0(\Delta_c, T_K)$. In fact, it has been proposed that $T_0 \approx \Delta_c$, provided $T_{max} >> T_0 \approx \Delta_c >> T_K$. In the same paper[34] a comparison of experimental data of the concentration dependence of T_{max} and T_0 for the *Au*Fe system supports this result. The Δ_c values (calculated from the measured T_{max} using $T_K = 0.19$ K in the L I formula 79) coincide with the spin-glass freezing temperatures T_0 as measured in the susceptibility in the concentration range from 0.1% to 1% (where the self-damping is negligibly small). The authors state, however, that the equality between the Δ_c and

TABLE I. Concentrations c, annealing temperatures T_a, mean-free-paths λ, temperatures of the maximum in $\Delta\rho$, T_{max}, and values for the freezing temperatures T_0 as determined by different theories: L I (Ref. 16): $T_0 = \Delta_c$; Δ_c has been calculated by Eq. (79) in L I taking $T_K = 0.19$ K and the measured T_{max} values. L II (Ref. 18): T_0 has been calculated by Eq. (5) in L II ($\beta = 1, A/k = 502$) as a function of the mfp λ. F I, F II (Refs. 19–21): The $T_0(\lambda)$ values are normalized to the T_0 values of the bulk samples. F I: this case should hold for spin-glasses with small concentrations of magnetic impurities and with interactions of sufficiently short range. $T_0 \propto c$. F II: this case holds for spin-glasses with uncorrelated spins and interactions. $T_0 \propto c^{1/2}$.

c	T_a[K]	λ[Å]	T_{max}[K]	T_0[K]			
				L I	L II	F I	F II
0.24 at%	9	43			2.37	1.32	2.48
	50	50	5.4	2.45	2.42	1.40	2.5
	150	70	6.0	2.66	2.50	1.59	2.57
	220	88	6.5	2.83	2.54	1.72	2.60
	300	195	8.7	3.56	2.65	2.19	2.66
	bulk	460	10.5	4.13	2.7	2.7	2.7
0.6 at.%	9	36	13.5	5.05	5.50	3.45	5.68
	50	40	14.5	5.35	5.58	3.62	5.73
	120	47	14.8	5.50	5.69	3.87	5.82
	220	60	16.8	6.03	5.83	4.25	5.93
	300	112	21.0	7.23	6.07	5.26	6.12
	470	135	23.0	7.79	6.12	5.57	6.16
	bulk	191	(25.0)	8.35	6.2	6.2	6.2
6.0 at.%	9	13.3			22.6	19.3	22.4
	50	13.8			22.8	19.9	22.5
	150	14.9			23.2	21.1	23.2
	220	15.9			23.6	21.8	23.4
	300	17.8			24.1	23.4	24.2
	bulk	19.2			24.5	24.5	24.5

T_0 values might be somewhat fortuitous.

Following this result, the variation of T_0 with the mfp can be directly deduced for the films. The data are given in Table I, column L I. They have been calculated using the experimentally determined values for T_{max} and a Kondo temperature $T_K = 0.19$ K. Possible changes of T_K with λ are disregarded in this analysis. The analysis shows that the variation of T_0 with λ is appreciable, e.g., for the 0.24% alloy of the order of 75% between the film with the highest degree of disorder (shortest λ) and the bulk. Furthermore, it should be mentioned that the freezing temperatures for the bulk turn out to be rather high compared to those determined from susceptibility measurements.

We think, however, that the variation of T_0 with λ cannot be predicted correctly by L I. One would not expect that a theory describing a physical property like T_{max}, which originates from a competition of the Kondo effect and the RKKY interaction, can allow a direct deduction of T_0, which is a pure spin-glass property.

In a further paper[18] (hereafter L II), Larsen calculated the concentration dependence of the spin-glass freezing temperature, also taking the damping of the RKKY interaction into account. Equation (5) in L II allows a direct calculation of the variation of T_0 with λ for our thin films, if one normalizes the data to the T_0 values of the bulk samples and uses $\beta = 1$, $A/k = 502$. These T_0 values are also given in Table I, column L II. One can see that in this case the change of T_0 with λ is small for all concentrations of the order of 10%.

Recently, a different theoretical approach on the basis of a generalized mean-field theory of an Ising spin-glass, taking into account correlations between the spins and their interactions has been given by Kinzel and Fisher.[19-21] Two limiting cases of the model are discussed:

(i) The spins and interactions are completely correlated, in which case $\chi(0) \to 0$ for $T \to 0$ and $T_0 \propto c$ (scaling). Case (i) corresponds to spin-glasses with small concentrations of magnetic impurities and with interactions of sufficiently short range. It can, however, probably not be realized experimentally.

(ii) Spins and interactions are uncorrelated. Here $\chi(0) = 0.64\chi(T_0)$—which is experimentally realistic—and $T_0 \propto c^{1/2}$, which holds for high concentrations. Case (ii) perhaps could also be fulfilled in amorphous spin-glasses or in the disordered films, in which the range of RKKY interactions is strongly reduced.

The authors also investigate the dependence of T_0 on the mfp λ taking into account the usual damping factor $e^{-r/\lambda}$. Their result allows a numerical calculation of the variation $T_0(\lambda)$ in the two limiting cases (i) and (ii). We use the λ values as measured in the resistivity of the films and normalize the data to the T_0 of the bulk samples. The results are given in Table I under the columns F I and F II. One can see that in case (ii)—which should hold for the disordered films—the overall variation of T_0 with λ is small, of the order of 10% for all concentrations, and comparable to L II. In case I—which is a nonrealizable limiting case—the variation $T_0(\lambda)$ is large, especially for low concentrations. We determine for the 0.24% alloy a change from $T_0 = 1.4$ K ($\lambda = 50$ Å) to $T_0 = 2.7$ K ($\lambda = 460$ Å) for the bulk sample. This is closer to the changes predicted by L I. For the 6-at.% alloy case F I predicts a change from $T_0 = 19.3$ K ($\lambda = 13.3$ Å) to $T_0 = 24.5$ K ($\lambda = 19.2$ Å). Although case (i)—complete correlation, i.e., totally ordered spins—should not hold here, there are early and recent experimental data by Korn[35,36] and Zibold and Korn[37] on the variation of T_0 in quench condensed films, and the change of T_0 during annealing. For example, in their recent measurement on a Au + 4-at.%-Fe alloy film[37] these authors have found a change from $T_0 = 12.5$ K in the quench condensed film to $T_0 = 17$ K in the annealed status ($T_a = 800\,^\circ$C). This change would better be described by the cases L I or F I, but it is unknown to what extent a rearrangement of nearest-neighbor Fe atoms during the annealing would influence the value of T_0 in films with impurity concentrations that high.

In conclusion, one can say that at present it is not predictable how much the spin-glass freezing temperature T_0 will change in low concentrated alloys, if the damping of the RKKY interaction is increased by shortening the mfp of the conduction electrons. It is, however, quite obvious that a spin-glass with a high concentration of magnetic impurities is less sensitive to additional nonmagnetic scattering of the conduction electrons than a dilute system. The relative degree of disorder which can be introduced into high concentrated thin-film systems is much lower and self-damping is more important. One would expect therefore the bigger changes of T_0 with λ to occur in the lower concentrated spin-glass films, as, e.g., predicted by the theory of Kinzel and Fischer,[19] case (i).

C. Low-temperature behavior of the resistivity

In the following, the temperature dependence of the resistivity in the spin-glass region, i.e., for $T < T_{max}$, and its variation with the mfp will be analyzed. Unfortunately, to date no comprehensive theoretical description of all the effects observed experimentally does exist. Thus, we will discuss $\Delta\rho(T)$ in terms of different power laws in T, and compare it to available theoretical models for each range separately.

1. Linear-T region

Figure 2 shows the low-temperature part of the resistivity for the two more dilute alloy films with different λ plotted versus temperature on a linear scale, together with the data for the corresponding bulk samples. For comparison with the 0.6% film we

use the 0.5% bulk sample of Mydosh *et al.*[28] Two general features can be seen from the graph. Firstly, the slope of the curves increases with decreasing mfp and secondly, the width of the linear range increases with increasing magnetic impurity concentration. This width, however, does not change very much with the variation of λ.

At low-temperature linear T dependence of the resistivity in dilute magnetic alloys has been predicted theoretically by Harrison and Klein.[38] They introduced a short-range interaction between magnetic impurities within the framework of a molecular field theory. The authors assume each individual spin to be submitted to a molecular field H_n, the values of H_n

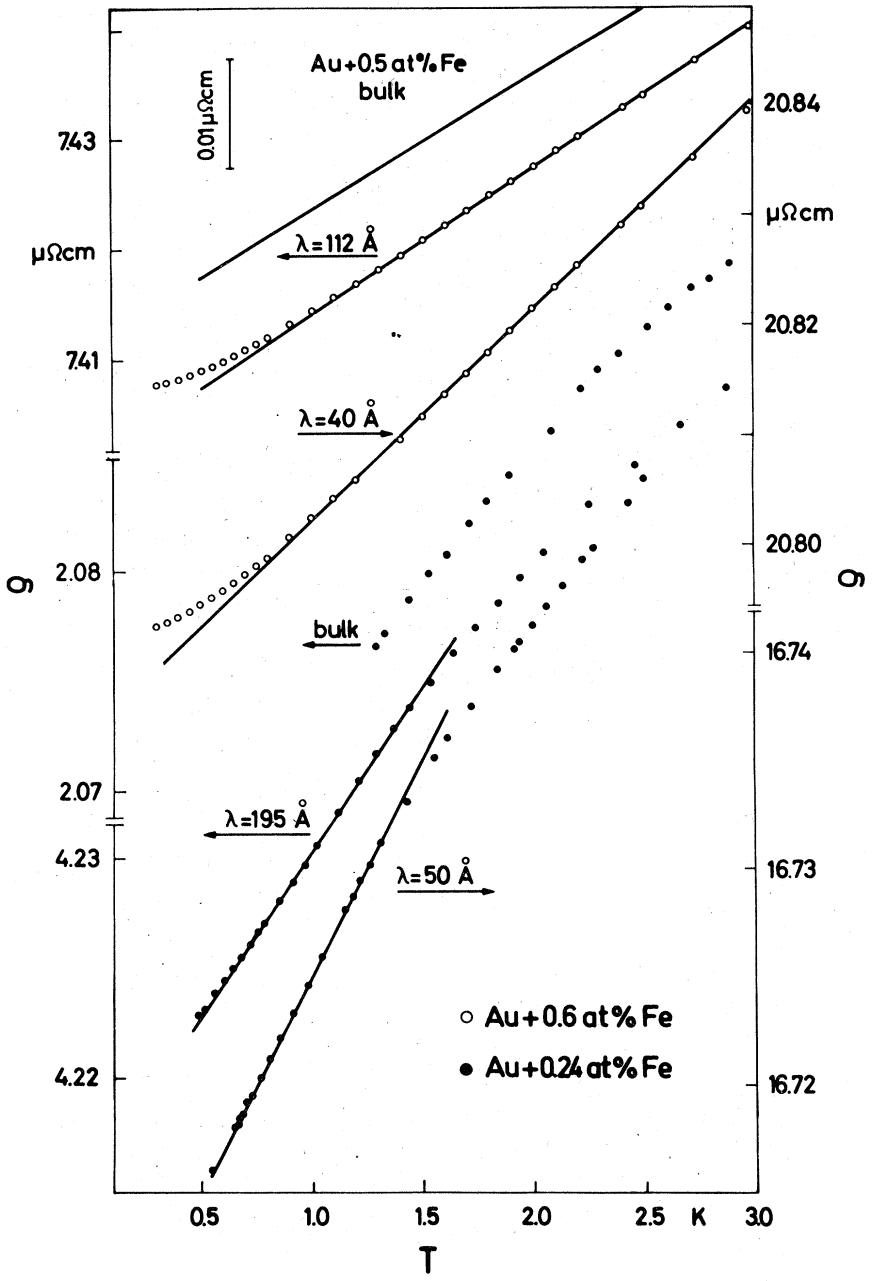

FIG. 2. Resistivity ρ vs temperature T for two different *Au*Fe alloys. The existence of a linear-T regime is demonstrated by the solid lines. The uppermost curve shows the temperature dependence of the resistivity for a Au + 0.5-at. at.% Fe bulk sample measured by Mydosh *et al.* (Ref. 9)], which has been chosen for comparison.

corresponding to a random variable resulting from the random distribution of the magnetic impurities. From the distribution $P(H)$ the different properties of the system have been deduced. Especially, it has been shown that the distribution function $P(H)$ derived within an Ising model[38–40] has Lorentzian character, with $P(H=0)$ being infinite and inversely proportional to the impurity concentration $P(H=0) \propto 1/c$. This scaling behavior of $P(0)$ is a direct consequence of the $1/r^3$ variation of the RKKY interaction.[41] Within a perturbation treatment, Harrison and Klein[38] have shown that for $0 < T << T_{max}$ a linear temperature dependence of $\Delta\rho(T)$ exists, the slope $m = \partial(\Delta\rho)/\partial T$ being proportional to

$$m \propto SJ^2cP(0) \ ,$$

where S is the impurity spin, J is the exchange constant.

Taking into account $P(H=0) \propto 1/c$ it follows that m is concentration independent. By using the width Δ of the distribution $P(H)$ as determined experimentally in specific-heat measurements on *Au*Fe, the authors[38] have found by a numerical calculation $m = 8\,\mathrm{n\Omega\,cm/K}$. This corresponds rather well to $m = 13-14\,\mathrm{n\Omega\,cm/K}$ as derived from our results (Fig. 2) and other experiments.[30] The numerical calculation also confirmed the experimental result that the width of the T range increases with increasing impurity concentration.

The increase of m with decreasing λ can also be understood from this model. The weakening of the RKKY interaction strength by the damping factor $e^{-r/\lambda}$ results in an increase of the number of sites in zero field $cP(0)$. Therefore, the slope m in the linear T region increases with decreasing λ reaching values of 20 $\mathrm{n\Omega\,cm/K}$ for the highest degree of disorder (see Fig. 2).

Although the molecular field model seems to describe well the experimental situation, it should be mentioned that its application to spin-glasses is probably incorrect. Firstly, there are power laws in T of higher order observed in the resistivity at lower temperature (see below) which are not found by Harrison and Klein.[38] Secondly, the peak in the susceptibility at the spin-glass freezing temperature T_0, which is characteristic for spin-glasses, cannot be described at all within the molecular field picture.

We think a different explanation is more adequate. According to a theoretical work by Rivier and Adkins[13]—which will be discussed in detail below—the linear T range is just a transition region between T_{max} and the low-temperature spin-glass characteristic $T^{3/2}$ regime. Thus, the increase in slope with decreasing λ in the linear range is just a consequence of the shift in T_{max} with λ to lower temperatures.

As can be seen from Fig. 2, the position and extension of the linear T range do not change very much with the mfp. This result is interesting in relation to the shift of T_0 with λ, as discussed in the preceding chapter. Since T_0 lies in the linear T range, a small shift of this range with λ of about 0.1–0.2 K as observed in Fig. 2, would support more the models L II and F II (see Table I), where changes of T_0 of the same order of magnitude have been proposed. As long as there is no direct way to determine T_0 from resistivity measurements, this point has to be left open in the discussion.

A remark should be made concerning the temperature T_m of the maximum in the derivative of the resistivity, and its relation to T_0, as discussed by Ford and Mydosh.[9] These authors have found that within a concentration range $3.0 \leq c \leq 6$-at.%-Fe in Au the maximum $d\Delta\rho/dT$ coincided with T_0. A detailed analysis of the available data[42] on *Au*Fe, especially those at lower concentrations shows that this agreement is accidental. We find for the concentration dependence of T_m: $T_m \propto c^{1.15}$, and for T_0 above 1 at.%: $T_0 \propto c^{0.6}$. Within the concentration range $3 \leq c \leq 6$ at.% these two curves just cross.

In a recent paper[9] it has been argued that a shoulder can be present in $d\Delta\rho/dT$ if the magnetic clusters are sufficiently large and their average radius r_0 is larger than the electronic mfp λ. This shoulder can be roughly correlated with T_0. For binary alloys $r_0 > \lambda$ should hold only at high impurity concentrations, and indeed in, e.g., Cu + 6.3- and 10-at.% Mn a shoulder in the derivative curves of the resistivity has been observed.[9] In bulk *Au*Fe no shoulder has been found at all.

In the films the situation is different, since the condition $r_0 > \lambda$ can be fulfilled already at low impurity concentrations just by reducing the mfp by quench condensation. Figure 3 shows the result for the 0.24% alloy film, where computer calculated values of $d\Delta\rho/dT$ are plotted versus temperature with λ as the parameter. Besides the maximum, always present at $T \approx 1$ K for all λ, the development of the shoulder at $T \approx 2.6K \approx T_0$ and its shift with increasing λ is clearly visible. Similar results are obtained for the other samples. However, as long as details about the freezing process are still unknown, it is difficult to get information about it from transport properties.

2. $T^{3/2}$ and T^2 region

The $T^{3/2}$ temperature dependence of the resistivity, observed for temperatures below the linear T region, is supposed to be typical for spin-glasses. Figure 4 gives an example for the 0.6-at.% film and shows that the slope—as in the T range—increases with decreasing mfp. The origin of the $T^{3/2}$ power law has been discussed within the spin diffusion theory by Rivier and Adkins.[13] They explained the low-temperature resistive behavior in terms of the scattering of conduc-

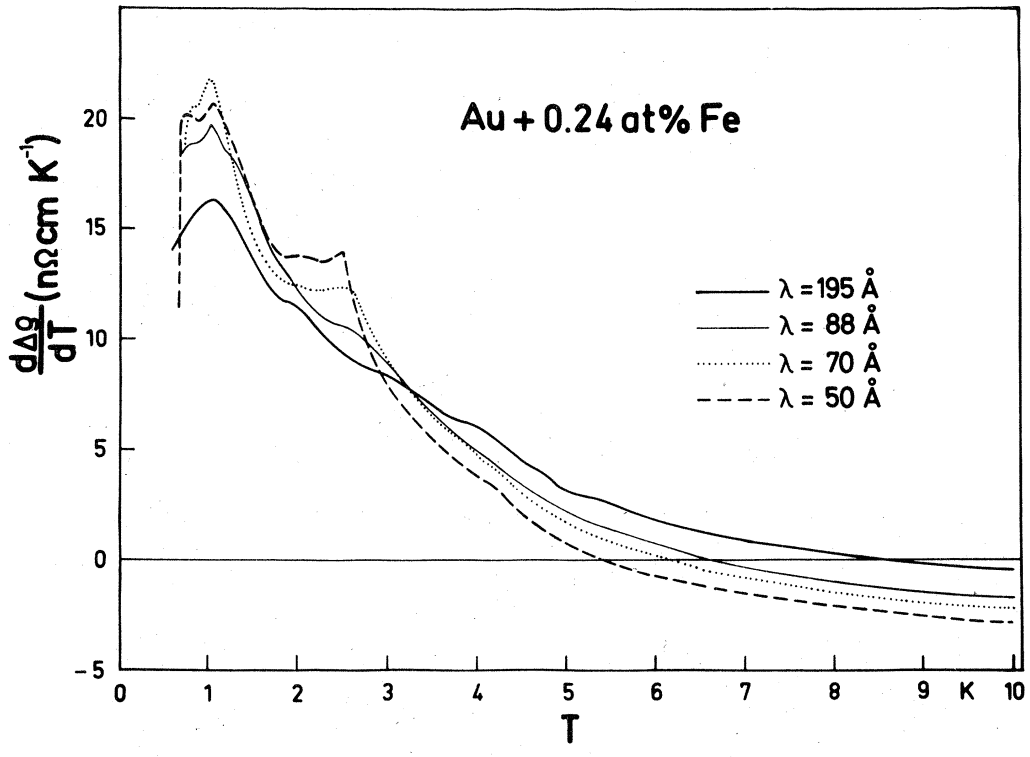

FIG. 3. Temperature dependence of the temperature derivative of the impurity resistivity $d\Delta\rho/dT$ for the Au + 0.24 at.% Fe film annealed at different temperatures. In addition to the maxima whose position does not change with the mfp, the development of a shoulder around $T = T_0 = 2.6$ K can be seen with falling mfp λ.

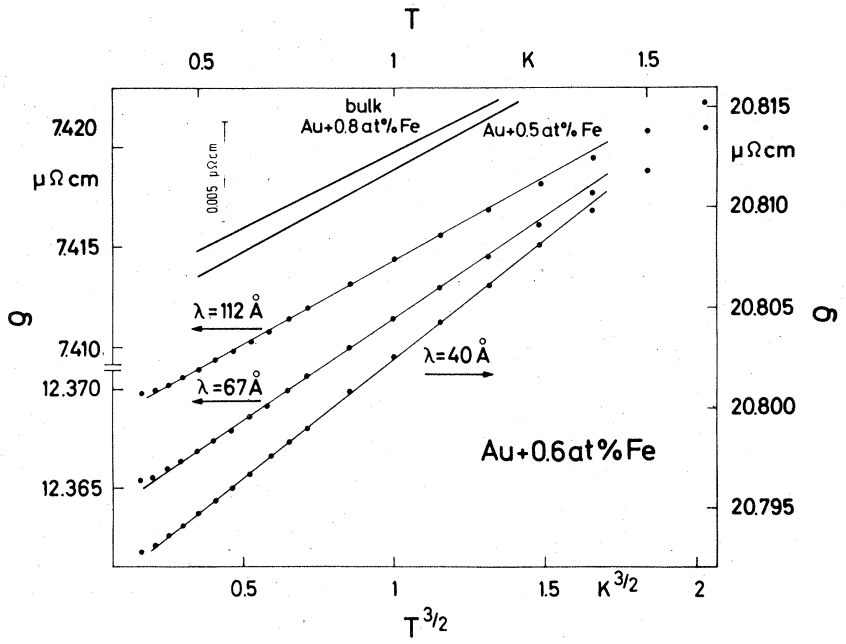

FIG. 4. Resistivity ρ as a function of $T^{3/2}$ after three annealing steps for the Au + 0.6 at.% Fe film. The $T^{3/2}$ regimes are illustrated by thin solid lines. For comparison the temperature dependence of the resistivity for two bulk samples (Au + 0.5 at.% Fe and Au + 0.8 at.% Fe) are shown by the thick solid lines.

tion electrons by long-wavelength spin diffusion modes. The model is restricted to a supercooled paramagnet, i.e., a spin-glass with negligible short-range order. The coefficient of the $T^{3/2}$ law, A in the Rivier and Adkins theory, depends weakly on the impurity concentration. Mydosh *et al.*[28] have found experimentally $A \propto -\ln c$ or a dependence like $A \propto c^{-1/5}$ describing the results equally well.

The c dependence of the A values of the films is qualitatively the same as in the bulk, but our absolute values of A are higher and more consistent with the data for a dilute bulk 0.03-at.% *Au*Fe alloy for which we have recently measured $A = 15\ \mathrm{n\Omega\,cm/K^{3/2}}$ for temperatures below 0.3 K.

According to Rivier and Adkins[13] the slope of the $T^{3/2}$ region A is also a function of the electronic mfp. For constant impurity concentration they find $A \propto \ln(\lambda/a)^{3/2}/\lambda^3$, where a is the lattice constant. For $\lambda >> a$ this corresponds approximately to $A \propto 1/\lambda^3$. As can be seen in Table II for the 0.6-at.% alloy, A indeed decreases appreciably with increasing mfp. Unfortunately, for the 0.24-at.% alloy only the A coefficient after annealing of the film to 300 K could be determined. Thus, for a quantitative determination of the λ dependence of A more experimental data are necessary.

In the Rivier and Adkins[13] theory the low-temperature $T^{3/2}$ behavior of the resistivity depends critically on the availability of spin diffusion modes with very long wavelength. If these modes are no longer operative, e.g., due to spin relaxation of the conduction electrons by additional imperfections, then a T^2 dependence of $\Delta\rho$ is expected at low temperatures (as in a least-squares-fit (LSF) theory). Clearly, this has to be the case in the quench condensed thin films, where a high amount of lattice defects is additionally introduced by the evaporation process. With increasing annealing the concentration of these defects is progressively reduced. As a consequence, the T^2 region should be less pronounced. These predictions are confirmed by our measurements, as shown in Fig. 5, where the impurity resistivity is plotted as a function of T^2 for the 6% sample. While for the highly disordered film with $\lambda = 13.3$ Å the T^2 region extends from 1 to 3 K, the annealed film with $\lambda = 17.8$ Å shows a T^2 law only between 1 and 1.5 K.

TABLE II. Annealing temperatures T_a, mean-free-paths λ, and coefficients of the $T^{3/2}$ law, A, for the film Au + 0.6-at.% Fe.

c	T_a [K]	λ [Å]	A [$\mathrm{n\Omega\,cm/K^{3/2}}$]
0.6 at%	9	36	11.9
	50	40	11.9
	120	47	11.7
	170	53	11.8
	220	60	11.3
	260	67	10.3
	300	112	8.4
	470	135	7.2

So far, a T^2 dependence has been found in lower concentrated bulk alloys of *Cu*Mn, *Au*Fe, and *Au*Mn ($0.075 \leq c \leq 1$ at.% in all systems) by Laborde[30] for temperatures below $T/c < 0.25$ K/at.%, a temperature range not reached in the earlier work of Mydosh *et al.*[28] on *Au*Fe. In higher concentrated alloys of *Au*Cr (7.9 at.%; 10.6 at.%) a T^2 temperature dependence has been measured above 1.5 K by Ford and Mydosh.[9] Our results on the thin films demonstrate that with increasing mfp and decreasing concentration the extension of the T^2 range gets shorter and shorter, and the interval shifts to lower and lower temperatures. We think that the occurrence of a T^2 dependence in the resistivity at very low temperatures is a general feature of all spin glasses, although its existence might be difficult to verify, especially for low concentrated bulk alloys.

3. Resistivity minimum

Figure 5 shows that at very low temperatures a minimum occurs in the resistivity of the disordered 6-at.% *Au*Fe film. An indication of the presence of such a minimum has also been found for the quench condensed 0.6% film. The position and depth of the minimum clearly depend on the mfp of the conduction electrons. In Fig. 5 it shifts from $T_{\min} = 0.84$ K for $\lambda = 13.3$ Å to $T_{\min} = 0.65$ K for $\lambda = 13.8$ Å and for $\lambda = 17.8$ Å out of the range of measurement. Minima are normally not observed in bulk material, except when the impurity concentration is very high (very high self-damping) as in the Au + 22.2-, 28.3-at.% Cr alloys by Shiozaki *et al.*[43] The minimum could be understood as a result of the Kondo effect of residual magnetic impurities, which—because of the short λ—have "escaped" the RKKY coupling, and thus do not participate in the freezing process. The occurrence of isolated impurities can, e.g., be understood from the statistics of an Ising model[44] (see also Sec. III C1). However, for a quantitative study of the temperature dependence of $\rho(T)$ below the minimum as well as the determination of $\rho(0)$ investigations in the mK range are necessary.

IV. SUMMARY

Within the present investigation it has been shown that the low-temperature resistivity behavior of the spin-glass can appreciably be changed if the mfp of the conduction electrons of the matrix is changed. This

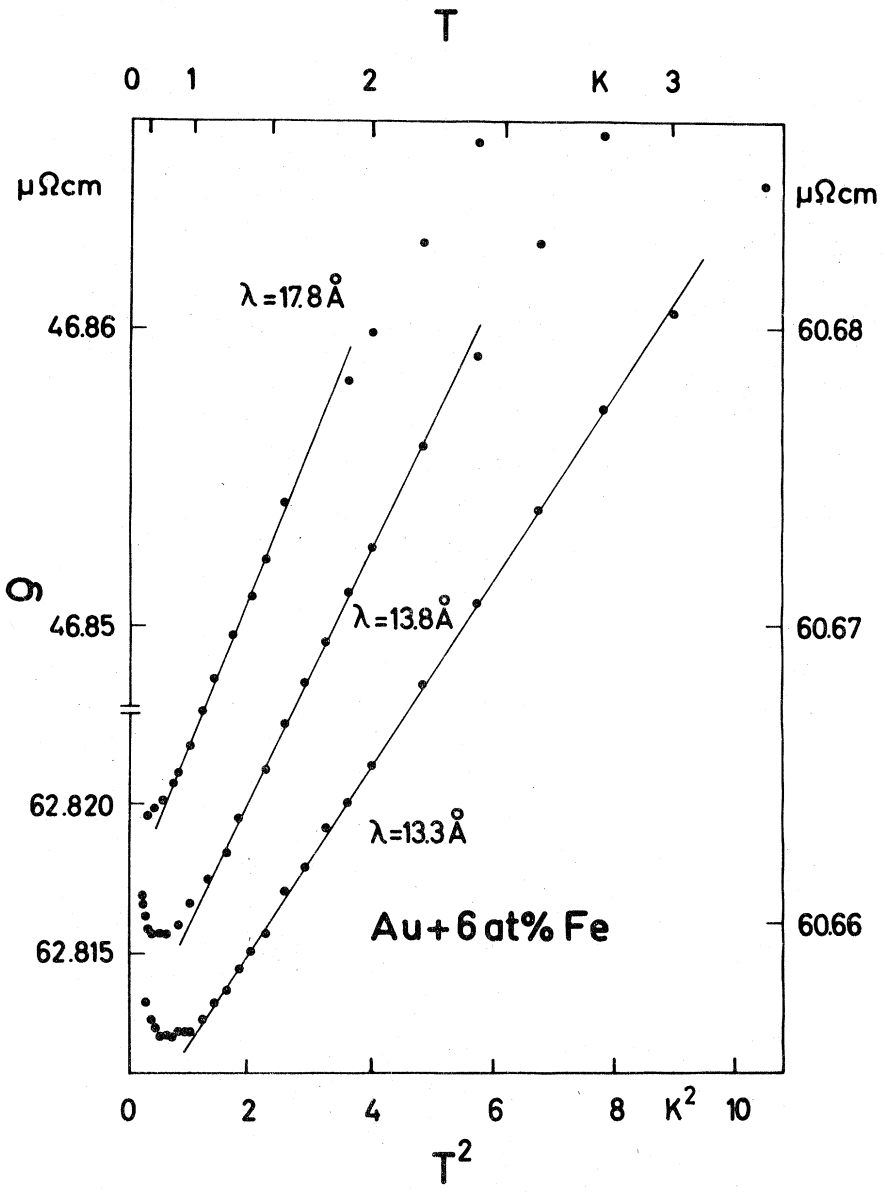

FIG. 5. Resistivity ρ as a function of T^2 after three annealing steps for the film Au + 6.0 at.% Fe. The solid lines show the T^2 dependence. Minima are clearly visible for the two lower curves. T_{min} decreases with increasing mfp λ.

variation of λ can be achieved by introducing nonmagnetic scattering centers into the alloys. We have used the method of quench condensation of thin films, with consecutive annealing at higher temperatures. A shortening of the mfp of the conduction electrons results in a damping of the amplitude of the RKKY interaction—expressed by the de Gennes factor $e^{-r/\lambda}$—and thus with decreasing λ the impurity-impurity interaction strength is progressively reduced. In the resistivity dependence of the *Au*Fe films this has the following results.

(i) There is higher spin scattering in the region $T > T_{max}$, where the Kondo effect governs the resistivity behavior. The impurities are "decoupled" to such an extent that even in a relatively concentrated Au + 0.24-at.%-Fe film the "one-impurity limit" is approached for $T > 15$ K.

(ii) We observe a shift of the maximum temperature T_{max} to lower values when λ is reduced. From the dependence $T_{max}(\lambda)$ a shift of the spin-glass freez-

ing temperature T_0 with λ has been deduced by applying different theoretical models. Variations of T_0 between 10% and 100% are predicted, depending on the model used. From our resistivity measurements, however, we cannot decide which model is adequate.

(iii) We obtain changes of the position, width, and slope of the different power laws, describing the resistivity for $T < T_{max}$. There is always a T^2 dependence present in the resistivity of a spin glass for $T \ll T_{max}$, when the mfp is short. This is well understood within the spin diffusion theory of Rivier and Adkins.[13]

(iv) We find the occurrence of a minimum in the resistivity of the films below the T^2 region, when λ is very short. The minimum can probably be understood as the Kondo effect of residual magnetic impurities.

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