

## Magnetic Interactions in a Weak Moment System: $Au-Co$ <sup>†</sup>

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Bulk magnetic measurements have been made on  $Au-Co$  (5 and 7 at. %) between 1.5 and 300 °K. The high-temperature susceptibility is compared with that in dilute samples, from which it is concluded that interactions among the Co atoms strengthen the virtual Co moments. This conclusion is confirmed by the low-temperature properties which indicate ordering, e.g., susceptibility maxima, hysteretic magnetization curves, and time- and temperature- dependent remanences. Low-temperature measurements are presented on a strong moment alloy  $Au-Fe$  1 at. %. The low-temperature ordered-state properties of the  $Au-Co$  and  $Au-Fe$  specimens are qualitatively, and semiquantitatively, similar.

### I. INTRODUCTION

Experiments to investigate the existence and properties of a local moment on an isolated magnetic impurity must be done on specimens sufficiently dilute to make impurity-impurity interactions negligible.<sup>1</sup> Other experiments have focused on these interactions by studying concentrated alloys and, in particular, observing effects of ordering of the moments at low temperature.<sup>2-6</sup> In the case of systems which possess a local moment in the Friedel-Anderson<sup>7,8</sup> sense, such as  $Cu-Mn$  or  $Au-Fe$ , the existence of a local moment on the impurity can be presupposed conceptually, and models of the interactions of these moments can be constructed to account for the observations in concentrated alloys.<sup>5,9</sup> Other systems, e.g.,  $Au-V$  and  $Au-Co$ , behave "magnetically" in the dilute regime, in the sense of showing a markedly temperature-dependent susceptibility<sup>10,11</sup> and resistance minimum phenomenon,<sup>12,13</sup> yet one must consider the moments "weak" in the sense of a weaker temperature dependence indicated by a large negative  $\theta$  in a fit of the high-temperature susceptibility to a Curie-Weiss law,  $\chi = C/(T - \theta)$ . Also the interactions of the impurities are weaker as seen in the absence of the onset of order in a 1 at. % alloy.<sup>6</sup> The validity of possible equivalence of interpreting these systems as ones of high Kondo temperature<sup>14</sup> or short spin-fluctuation lifetime<sup>15</sup> has not been theoretically clarified at present.

Intuitively, one expects that in these weak moment systems the previous separation of the *existence* (dilute property) and *interaction* of the moments cannot be made. For instance, in the localized-spin-fluctuation picture, one would expect interactions with a sufficiently nearby impurity neighbor to affect  $\tau_{SF}$ . This effect has been most clearly demonstrated in  $Au-V$ <sup>10,16</sup> where it is found by susceptibility and NMR measurements that near-neighbor V pairs demagnetize one another. On the other hand, ordering effects have been seen in concentrated (2-4-at. %)  $Au-Co$ ,<sup>17</sup> indicative of stabilization

of the moments due to the presence of nearby Co atoms. A similar onset of magnetic order at large concentration (compared with those required in strong moment dilute alloys) has been seen in  $Rh-Fe$ .<sup>18</sup>

Further work has been undertaken on the magnetic properties of  $Au-Co$  well into the region where magnetic ordering occurs (>4-at. % Co). The high-temperature measurements are compared with Hildebrand's<sup>11</sup> measurements on dilute specimens. Low-temperature measurements have also been made on  $Au-Fe$  to compare the magnetic properties of the ordered state in a weak ( $Au-Co$ ) and strong ( $Au-Fe$ ) magnetic moment system.

### II. EXPERIMENTAL PROCEDURE

The samples were prepared in an argon arc furnace using weighed amounts of 99.999% pure Au rod and Co (or Fe) wire, all being obtained from Koch-Light, Ltd. The buttons were turned and remelted several times to achieve mixing. They were then swaged and rolled into a block and homogenized *in vacuo* at 1000 °C for several days, which was followed by quenching into water. The sample was then rolled, filed, and ground into the desired cylindrical form, reannealed at 1000 °C *in vacuo* about a day, and again quenched. The specimens were quantitatively analyzed by atomic absorption and by conventional chemical methods. The chemical analysis agreed well with the weighed input amounts, 5.0 and 6.8 at. %, while the atomic absorption results were about 0.4 at. % lower, well outside the estimated respective errors. Since the weight loss during melting was small and the chemical results considered more accurate, the compositions of the 5- and 7-at. % Co alloys will be taken as (5.0 ± 0.1) and (6.8 ± 0.1) at. %. Emission spectrographs showed Fe impurities of about 5 and 40 ppm in the 5- and 7-at. %  $Au-Co$  samples, respectively. The nominally 1% Fe specimen contains (0.96 ± 0.02)-at. % Fe. The large Fe impurity con-

tent in the 7% specimen will have a small effect ( $<0.1\%$ ) in its direct contribution to the magnetization since, as will be seen, the Co is almost as strongly magnetized with this concentration as is Fe in dilute solution in Au. The real problem is in evaluating the contribution of 40 ppm of Fe to the stabilization of the Co moments. The transition to strong magnetic behavior is so clear, however, that it is expected that the Fe impurities in the 7% alloy could only have a minor effect on this process.

Metallurgical and magnetic studies have been performed to study the precipitation of Co in Au-Co alloys in the vicinity of 5-at. % Co.<sup>19-21</sup> Electron micrographs<sup>21</sup> and magnetization measurements<sup>19</sup> show that precipitation of a Co-rich phase occurs with aging at an elevated temperature, but can be avoided with a quench from the region of solubility. Magnetization measurements on the present Au-Co specimens at 77°K showed no observable remanence, from which an upper limit of about 3 ppm can be set for the ferromagnetic Co content.

The possibility was considered that the low-temperature magnetic-ordering properties that were observed were due to incipient atomic separation, analogous to that in age-hardening Al-Cu alloys, since the magnetic behavior was first studied on samples that had been at room temperature for many days. To check whether such pre-precipitation effects were of significance in the low-temperature magnetic properties, the 7% Co specimen was reannealed at 1000°C, quenched, and run in the magnetometer the same day and several days following. All the properties found previously (low-temperature maximum in the susceptibility, hysteretic magnetization curves, low-temperature remanent effects, including time dependence) were reproduced, the only change being that all magnetizations were about 10% smaller after reannealing. A change of this order in the concentration would be obvious, for instance, in the ordering temperature (see Sec. IV), which was not observed to change. This decrease (which did not recover in the course of a week) is probably due to the complete loss of Co by internal oxidation in a thin surface layer, although faulty placement of the specimen or alteration of the nulling-coil calibration cannot be ruled out.

Finally, the quenching procedure was checked by slowly cooling a part of both concentration Au-Co ingots in order to induce precipitation. The Vickers hardness was found to be about 110. After reanneal and fast quench into water, the hardness was about 50, indicating a much more homogeneous solution.

The measurements were made with an apparatus to be described elsewhere.<sup>22</sup> A driven-tuning-fork arrangement provided sample motion along the field direction. The signal induced in a detecting

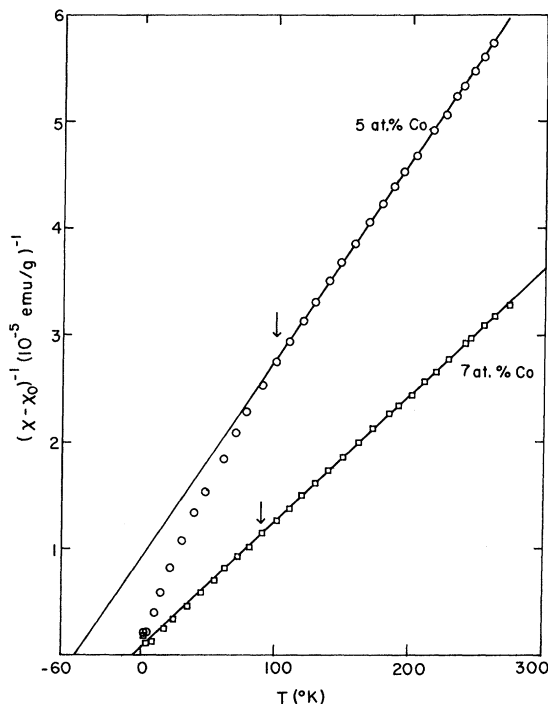


FIG. 1. Inverse susceptibility of the Au-Co specimens. The solid lines are a least-squares Curie-Weiss fit, using the data above a low-temperature cutoff indicated by the arrows.

coil was phase coherently detected. The current in a double layer coil wound around the specimen was adjusted to null the lock-in output, so that the measured quantity was this nulling-coil current, which is proportional to the specimen magnetization. Calibration was obtained by measurement of an identically shaped Pd specimen, whose susceptibility is known. The uncertainty in absolute accuracy was about 2%. The main source of relative error was synchronous vibration of the pickup coil, giving rise to a spurious signal on the order of  $5 \times 10^{-5}$  emu in a field of 4 kG. The absolute field error was about 1%. The temperature was measured at low temperatures by a carbon resistance thermometer, calibrated against He<sup>4</sup> vapor pressure and by a copper resistance thermometer at higher temperature. The maximum temperature error was found in the region 5–30°K and was estimated to be about 2%.

### III. HIGH-TEMPERATURE SUSCEPTIBILITY

In Fig. 1, the high-temperature susceptibility of the 5- and 7-at. % Au-Co data is present in a form indicating the fit to a Curie-Weiss law. In the absence of evidence for any larger temperature-independent susceptibility,  $\chi_0$  was taken as the

TABLE I. Curie-Weiss parameters and susceptibility maximum of  $Au-Co$ .

	From Hildebrand				From this paper	
$c$ (at. %)	0.4	1.2	1.9	3.4	$5.0 \pm 0.1$	$6.8 \pm 0.1$
$\theta$ ( $^{\circ}K$ )	247	197	162	87	$49 \pm 3$	$7 \pm 3$
$\mu_{\text{eff}}$ ( $\mu_B$ )	4.59	4.59	4.59	4.28	$4.06 \pm 0.10$	$4.33 \pm 0.10$
$T_N$ ( $^{\circ}K$ )					$2.8 \pm 0.2$	$5.9 \pm 0.3$

susceptibility of pure Au,<sup>10</sup>  $\chi_0 = -1.37 \times 10^{-7}$  emu/g. A least-squares Curie-Weiss fit to the data has been made with a rather arbitrary low-temperature cutoff indicated in Fig. 1. The parameters obtained are given in Table I, which includes the same parameters for more dilute samples.<sup>11</sup> In the localized-spin-fluctuation model, the decrease in  $\theta$  was interpreted as an increased spin-fluctuation lifetime. That  $\mu_{\text{eff}}$  is nearly independent of concentration indicates that all the Co atoms maintain their magnetic character in the concentrated alloys, in sharp contrast with concentrated V in Au, where  $\mu_{\text{eff}}$  drops sharply,<sup>10</sup> indicating a mutual demagnetization of near V atoms. The goodness of the Curie-Weiss fit using  $\chi_{Au}$  as the only temperature-independent contribution indicates that the magnetic properties of  $Au-Co$  can be analyzed simply in terms of the "moments" of the Co atoms (and their interactions).

#### IV. LOW-TEMPERATURE ORDERING

The initial susceptibility of the  $Au-Co$  specimens went through a broad maximum at low temperatures, as shown in Fig. 2. This phenomenon, which has been observed in strong moment systems<sup>2,6</sup> and, at higher concentrations, in weak moment systems,<sup>17,18</sup> indicates the onset of an "antiferromagnetic" order. Because of the random environment of the magnetic impurities, and the oscillatory nature of their interaction [e.g., the Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism], the ordered state was expected to be a magnetic "glass," with no long-range order. The broadness of the maximum was thus due to the distribution of effective internal fields felt by different moments.

The existence of this ordered state presupposes *bona fide* moments on the Co atoms which can lock into an ordered state. The temperature of the maximum in the initial susceptibility  $T_N$  can be taken as the ordering temperature, which is thus an indication of the strength of the interactions among the local moments. In Fig. 3 are presented the currently available data on the ordering temperature of  $Au-Co$ . The ordering temperatures taken from the work of Le Guillerm *et al.*<sup>17</sup> are the "blocking"

temperatures, which are obtained by extrapolating the low-temperature saturated remanence to zero. This gives a somewhat lower estimate of the ordering temperature than  $T_N$  for two reasons. The extrapolations of the remanence of  $Au-Co$  and  $Au-Fe$  shown in Fig. 5 yield a temperature which is 80–90% of  $T_N$ . Furthermore, they used a linear extrapolation of the low-temperature remanence which gave a lower cutoff than the curve used in Fig. 5. Le Guillerm *et al.* found a much higher estimate of the ordering temperature from the maximum in the high-field differential susceptibility  $dM/dH$ . In this work, the maximum in the initial susceptibility, which is well defined in the vicinity of  $T_N$ , where the remanence is quite small, was used to determine the onset of order. It was felt that this is a better indicator because it avoids the possibility of inducing *bona fide* moments on some Co atoms by the large applied field rather than by internal interactions.  $T_N$  of Ref. 17 was used as the quantity most readily compared to the present  $T_N$ , as explained above. Ford *et al.*<sup>23</sup> saw a leveling off of the resistivity in a 3%  $Au-Co$  sample which foreshadows the onset of order below 2 $^{\circ}K$ . The Lutes-Schmit<sup>6</sup> upper bound was deduced from the complete lack of observed susceptibility maximum in a

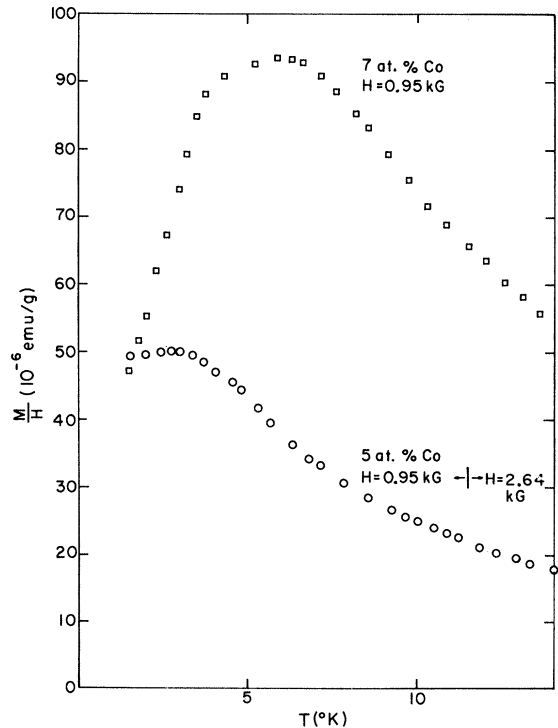


FIG. 2. Low-temperature initial susceptibility of the  $Au-Co$  specimens. The data below 4.2 $^{\circ}K$  were taken on cooling in the case of the 5% sample, and on warming, after cooling in zero field, in the 7% case.

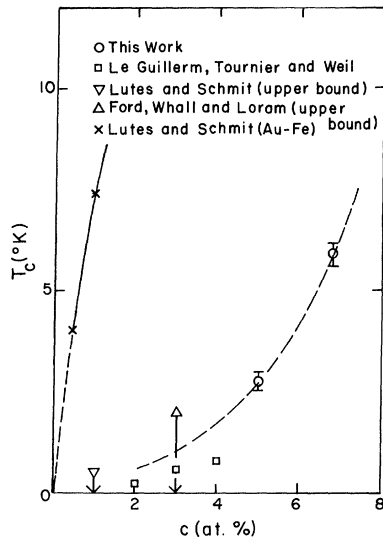


FIG. 3. Ordering temperature of  $Au-Co$  and  $Au-Fe$ . See Sec. IV for explanation of the data

1% sample down to  $0.5^\circ K$ .  $T_N$  for  $Au-Fe$  from Lutes and Schmit<sup>6</sup> is also shown for comparison.

Although the ordering temperature of  $Au-Co$  is not well defined by the divergent data available, it is clear that the system differs from the strong moment paradigm, e.g.,  $Au-Fe$ . The curvature of  $T_c$  versus  $c$  indicated by Fig. 3 substantiates the previous picture of Co-Co interactions strengthening the Co moments. That is, if  $T_c$  roughly proportional to  $c$  is taken as the strong moment paradigm, the stronger  $c$  dependence found in  $Au-Co$  arises from the additional concentration-dependent interactions needed to stabilize the Co moments. It is, of course, not clear whether there is a minimum Co concentration for ordering to take place at any temperature.

#### V. MAGNETIC HYSTERESIS

Below  $T_N$ , the magnetization curves of the  $Au-Co$  specimens show hysteresis. This phenomenon has been studied extensively in dilute  $Cu-Mn$  alloys<sup>3-5</sup> and also in Au-based alloys.<sup>6</sup> Magnetization curves of the 7% Co sample under various conditions are shown in Fig. 4. These closed curves were taken after cycling the field ( $\pm 8$  kG) under the particular field and thermal conditions. If the sample was cooled below the ordering temperature in zero field, a symmetric hysteresis curve was seen, such as the  $4.2^\circ K$  curve in Fig. 4, which shows a very small symmetric remanence. Similar, symmetric, zero-field-cooled hysteretic magnetizations were observed in  $Au-Co$  5% and  $Au-Fe$  1%, with increasing remanence with decreasing temperature.

When the sample was cooled below the ordering temperature in a large magnetic field, the magnetization curve was displaced upward if the direction that the field was cooled is taken as positive. This is illustrated in the  $1.6^\circ K$  curve in Fig. 4. This excess magnetization can be interpreted as a bias "frozen" into the array of moments as they order in the externally applied field.

#### VI. BEHAVIOR OF REMANENCE

Studies of  $Cu-Mn$  have shown<sup>4</sup> that it requires much less field to saturate the remanence by field cooling than by isothermal magnetization. This comparison was not done quantitatively in this  $Au-Co$  study because of the complicating time-dependent effects investigated (see Sec. VII). However, the  $1.6^\circ K$  field-cooled curve in Fig. 4 shows this effect in  $Au-Co$ . The positive displacement due to cooling in  $+8$  kG indicates the presence of a positive remanence which cannot be isothermally reversed by applying  $-8$  kG.

Since the strength of the interactions among the moments both determines the ordering temperature and provides the barrier against isothermal saturation of the remanence, it was possible to obtain a rough estimate of the field necessary for saturation by field cooling from Lutes and Schmit's measurements on  $Au-Fe$ , scaling the concentration to produce equal  $T_N$ . Their data for  $Au-Fe$  1% (the same

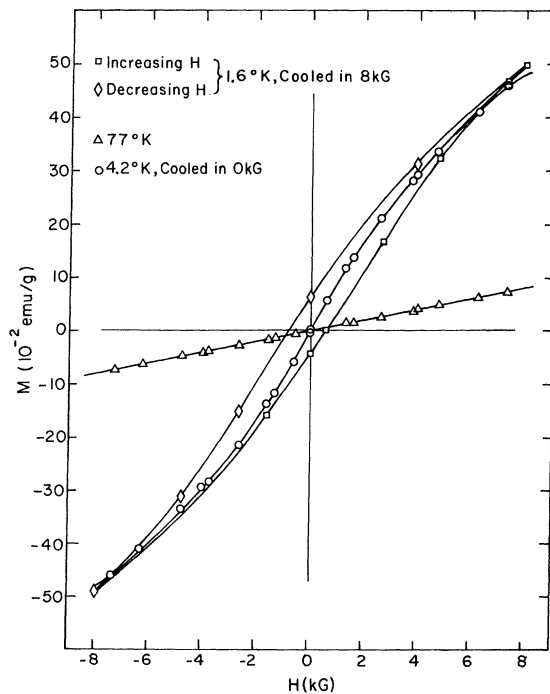


FIG. 4. Magnetization curves of  $Au-Co$  7%.

concentration used for the strong moment comparison here) at 1.33 °K (lower than any temperatures in the present experiments) shows a field of 6 kG to be sufficient to saturate the remanence by field cooling. Since the Au-Co 7% was easier to saturate (in terms of higher experimental temperatures and lower  $T_N$ ), the 8 kG available should suffice for field cooling saturation. The positive remanence values found after field cooling of the Au-Co 7% and Au-Fe 1% samples are shown in Fig. 5 (note the time dependence). Lutes and Schmit's<sup>6</sup> Au-Fe 1% data lie within the range shown in Fig. 5, down to 2 °K, but rise faster below 2 °K. The temperature of 6.3 °K for the extrapolation of  $\sigma_r$  to zero agrees well with their finding of 6.4 °K. As mentioned in Sec. IV, these temperatures are roughly 80–90% of  $T_N$ .

As previously suggested, the remanence of a field-cooled specimen can be divided into two parts:  $\sigma_H$ , that which can be isothermally reversed by the maximum field available (8 kG in the present case) and  $\sigma_s$ , that which keeps its original sign despite the application of -8 kG. Figure 4 shows that, for Au-Co 7% at 1.6 °K,

$$\sigma_H/\sigma_s = [\alpha_r(+)-\alpha_r(-)]/[\sigma_r(+)+\sigma_r(-)] \approx 6, \quad (1)$$

where  $\sigma_r(\pm)$  is the remanence after turning off  $\pm 8$  kG. Consider the temperature dependence of the negative remanence as the specimen is warmed in zero field, after waiting a long time after turning

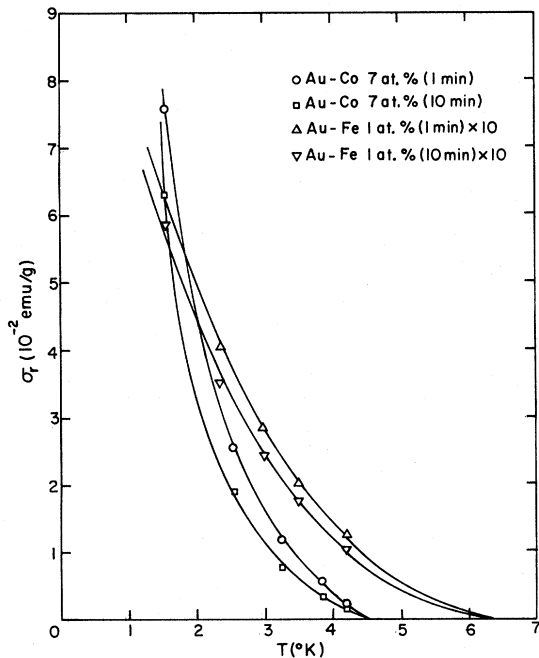


FIG. 5. Temperature dependence of the magnetic remanence, after successive field cooling in +7.33 kG.

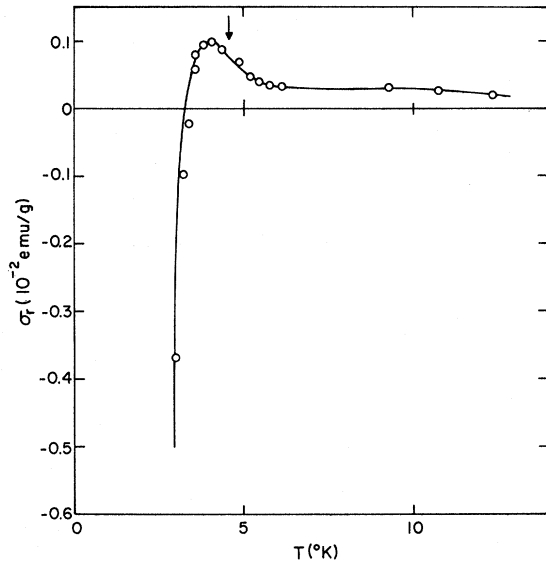


FIG. 6. Temperature dependence of the remanence of Au-Co 7%. The specimen was cooled in +7.33 kG and isothermally reversed by -8 kG at 1.5 °K.

off the field, so that the time dependence is slow (Sec. VII).  $\sigma_r(T) = \sigma_H(T) + \sigma_s(T)$ , where  $\sigma_H$  is negative. The high-temperature tail of such an experiment (expanded with respect to Fig. 5) is shown in Fig. 6. The reversal of sign was attributed to the fact that  $\sigma_s$  was expected to persist to higher temperatures than  $\sigma_H$ , since it was postulated to arise from Co moments experiencing strong interaction with other moments. The arrow in Fig. 6 indicates the temperature of the extrapolation of  $\sigma_r$  to zero in Fig. 5. Since  $\sigma_s$  is a small part of  $\sigma_r$  at low temperatures, this temperature is where  $\sigma_H \rightarrow 0$  and hence where  $\sigma_s$  dominates. Although not shown, this phenomenon was repeated in the Au-Co 7% sample after the reannealing mentioned in Sec. II. Moreover, a similar reversal of  $d\sigma_r/dT$  was observed in the Au-Fe 1% sample under the same conditions. That  $\sigma_r$  itself never went positive might have been due to a residual negative field, or the relatively larger influence of the sample holder residual magnetization since the magnetizations of the Au-Fe specimen is roughly 10% of that of the Au-Co.

## VII. TIME DEPENDENCE

The remanent magnetization of both Au-Co specimens and the Au-Fe specimen was observed to decay slowly after the field was removed. This phenomenon of "magnetic viscosity" has been observed in ferromagnetic alloys<sup>24</sup> and in Cu-Mn<sup>4,25</sup> and Au-Fe.<sup>26</sup> Using a model of domains which thermally overcome a barrier to reach equilibrium and as-

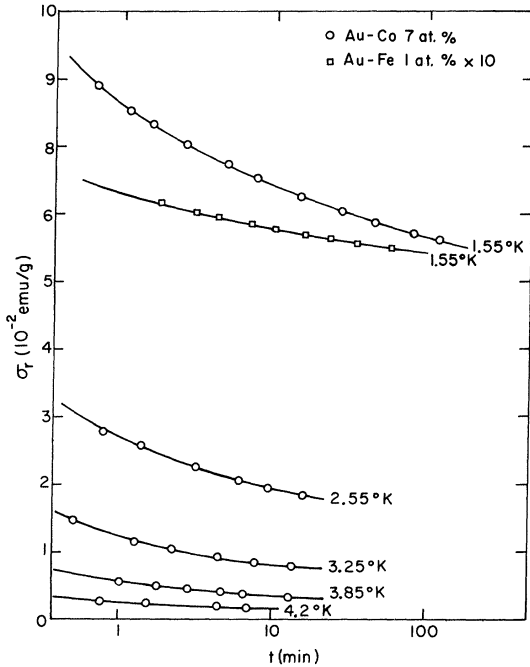


FIG. 7. Time dependence of the remanence after cooling in 7.33 kG. Time is measured from the middle of turning off the field, which takes about 15 sec.

suming a flat distribution of barrier heights, Street and Wooley have shown<sup>27</sup> that the magnetization varies linearly with  $\log_{10} t$  and  $T$  within certain approximations. Observations on *Au-Co* 7% and *Au-Fe* 1% are shown as a function of  $\log_{10} t$  in Fig. 7. The dependence on time is weaker than  $\log_{10} t$ . This contrasts with Lutes and Schmit's<sup>4</sup> results for *Cu-Mn*, where a stronger initial time dependence (nearly exponential) becomes logarithmic at long times.

Néel<sup>28</sup> has derived a relationship between the time and temperature dependence of the magnetization of fine grained ferromagnets  $(\partial\sigma_r/\partial \ln t)/(\partial\sigma_r/\partial T) = R$ . His expression contains a term which he estimates as about 30% for ferromagnets at room temperature and which goes to zero at  $T$ , so it will be ignored for the present analysis. Néel's expression reduces to

$$R = T/(Q + \ln \tau), \quad (2)$$

where  $Q \approx 22$ , and  $\tau$  is the measurement time in seconds. The data in Fig. 7 do not show the quite nearly linear dependence on  $\log_{10} t$  predicted by Néel's theory. Nevertheless, to compare the magnitudes of the observed time and temperature dependences with this theory, the expression  $R/T$  has been evaluated from the slopes of the fits to the data in Figs. 5 and 7 at 1 and 10 min after the field was turned off, and is tabulated in Table II. Néel's

expression  $Q + \ln \tau$  equals 26 and 28 for a  $\tau$  of 1 and 10 min, respectively. Correspondingly from Table II, taking a simple average of the different temperature *Au-Co* data, respective values for  $R/T$  of 21 and 36 for *Au-Co* and 14 and 27 for *Au-Fe* are obtained. The similar magnitudes obtained for these "magnetic glass" systems as for the fine grained ferromagnets indicate a similar time scale for the remanence to decay by reversal of some kind of domains.

## VIII. CONCLUSIONS

The magnetic properties of *Au-Co* alloys well into the interacting concentration region have been studied. Low-temperature ordering effects, i. e., susceptibility maximum, hysteresis, and time dependent remanence, have been observed. For comparison, a specimen of a strong moment system, *Au-Fe*, has been examined. Qualitative, and semi-quantitative, similarity of the magnetic properties in the ordered state was found. The concentration of the comparison *Au-Fe* sample was chosen to give roughly the same  $T_N$  as the *Au-Co* 7% specimen, which was most extensively studied. It appears that *Au-Co* requires a large concentration to achieve the mean interaction strength of a dilute *Au-Fe* sample, but when this is achieved, all the macroscopic magnetic properties are quite similar.

In the dilute regime, Co in Au is weakly magnetized, forming a virtual bound state which is unmagnetized in the Friedel-Anderson sense, but is capable of undergoing spin fluctuations, which give rise, at high temperatures, to a Curie-Weiss susceptibility with a large  $\theta$ . Interactions of the Co atoms stabilize their moments and, with large enough concentration, stabilize the moments sufficiently so that the magnetic properties become very similar to the strong moment *Au-Fe* system. It is interesting to consider the significance of the fact that  $p_{\text{eff}}$  stays nearly constant (Table I) across both these regimes. This can be interpreted to mean

TABLE II. Ratios of time and temperature dependence of the remanence  $R = (\partial\sigma_r/\partial \ln t)/(\partial\sigma_r/\partial T)$ .

Specimen	<i>Au-Fe</i>		<i>Au-Co</i> 7 at. %			
	1 at. %					
Temperature (°K)	1.55	1.55	2.55	3.25	3.85	4.2
$R$ (1 min) (°K)	0.11	0.073	0.18	0.21	0.15	0.08
$R$ (10 min) (°K)	0.064	0.035	0.12	0.091	0.10	0.062
$R$ (1)/ $T$	0.072	0.047	0.072	0.065	0.039	0.019
$R$ (10)/ $T$	0.037	0.023	0.047	0.028	0.026	0.015

that when the localized spin fluctuation forms momentarily, it self-consistently assumes the "full" moment, i. e., complete splitting.<sup>29</sup>

The transition between these regimes, i. e., the very-low-temperature behavior for small concentrations, is not clarified at present. (See Fig. 3.) A phenomenological discrete neighborhood model, which Tournier and Blandin<sup>30</sup> have applied to a similar Cu-Co system, might clarify this process of strengthening the Co moments.

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