

this observation should not be given too much weight, it does suggest that the magnitudes obtained for  $c^*$  are physically reasonable.

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### INTERACTION EFFECTS ON THE APPEARANCE OF A MAGNETIC MOMENT ON Co, THROUGH NUCLEAR SPECIFIC-HEAT MEASUREMENTS ON Au:Co ALLOYS\*

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The specific heat of Au:Co alloys down to 0.02 K is reported. The nuclear term ( $AT^{-2}$ ) is due to the magnetic Co atoms which are found to be those included in groups of three and more neighbors. The low-temperature linear term ( $\gamma T$ ) is the sum of the contributions of the magnetic Co groups, nonmagnetic pairs, and isolated atoms. We estimate fluctuation temperatures of 50 K for the pairs and 700 K for the isolated atoms.

The available data on the Au:Co system show that at low concentrations the alloys are nonmagnetic while phenomena which are generally considered as characteristic of the existence of a localized magnetic moment on the impurity are found at higher concentrations of Co.

Firstly, the resistivity shows a flat  $\rho(T)$  behavior for the low concentrations, but a large minimum is observed for concentrations over 0.1%.<sup>1</sup>

Secondly, the magnetization<sup>2</sup> is of Pauli type and remains small compared with alloys of the Cu:Mn type for the low concentrations, but for concentrations over 1% a remanent magnetization and a susceptibility maximum are observed.

Thirdly, above 1 K, specific-heat measurements<sup>3</sup> show an electronic term, concentration dependent, for concentrations below 3%, but for increasing concentrations a tendency towards a concentration-independent behavior is observed. Similar remarks can be made for the Cu:Co<sup>4</sup> system. In the early classification of Blandin and Friedel this has been considered a critical

case, i.e., one for which the criterion for magnetism is almost realized:  $U\rho(E_F)-1 \sim 0$ .<sup>5</sup> [ $\rho(E)$  is the density of states in the unsplit virtual bound state for one spin direction.]

The theory of localized spin fluctuations<sup>6</sup> has given rise to the notion of a fluctuation temperature<sup>7</sup>  $T_{SF}$  [ $1/KT_{SF} = \Pi\rho(E_F)/(1-U\rho(E_F))$ ] which would separate a nonmagnetic state (well below  $T_{SF}$ ) and a state where the impurity would exhibit all the properties of a magnetic impurity.<sup>8</sup> Caroli has shown  $\rho(E_F)$  to be sensitive to the interactions with other impurities.<sup>9</sup> Thus the fluctuation temperature should depend on the interaction; Lederer has shown that the susceptibility is different for an isolated impurity and an interacting pair.<sup>10</sup> Such an analysis is particularly convenient in the case of Co [where  $1-U\rho(E_F) \sim 0$ ] since a moderate interaction may be sufficient to make  $T_{SF}$  change by several orders of magnitude. Recently Tournier and Blandin<sup>4</sup> interpreted magnetic results on the Cu:Co system with this argument, attributing a lower fluctuation temperature to the groups of two atoms than

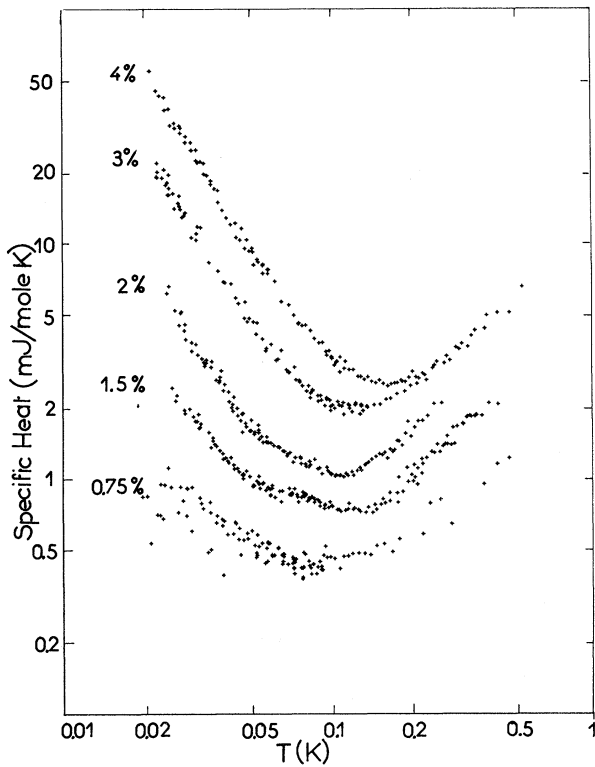


FIG. 1. Measured specific heat of Au:Co alloys versus temperature for different Co concentrations.

to the isolated impurities and a localized moment to the Co atoms involved in groups of three and more atoms.

We report here specific-heat measurements between 0.02 and 0.3 K on Au:Co alloys. The nuclear moment of Co being very large ( $4.6\mu_N$ ), the hyperfine  $T^{-2}$  contribution will be mainly produced by the effective field  $H_{eff}$  of the ordered magnetic Co atoms on their own nucleus and hence will be proportional to the concentration  $c_m$  of magnetic impurities. Then the hyperfine contribution allows one to enumerate the magnetic impurities. In addition, the term linear in temperature will provide some indication about the ordering of the magnetic impurities, and the contributions of the nonmagnetic atoms.

The samples used for this experiment were prepared from 99.999% Au and 99.999% Co. The constituents were melted in alumina crucibles under an atmosphere of purified hydrogen, and held 4 h at 1150°C. After removal of the crucible, the alloys (diam 7 mm,  $l \sim 20$  mm) were annealed 24 h at 950°C and quenched in a flow of gaseous hydrogen of 120 bars. This procedure gives reproducible results and is more efficient than quenching in water. After quenching, the

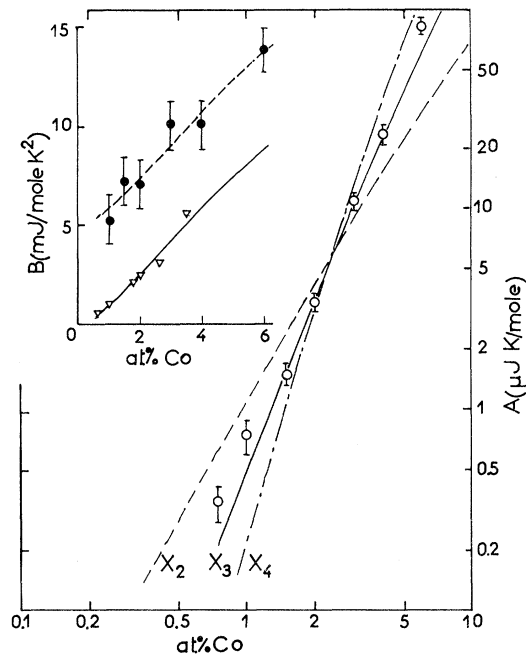


FIG. 2. The  $A$  coefficient of the  $T^{-2}$  term of  $C_p$  versus percent cobalt (for the 0.75, 1, 1.5, 2, 3, 4, and 6 at.% we find, respectively, 0.35, 0.75, 1.5, 3.4, 11.0, 23.4, and 82.3  $\mu\text{J K/mole}$ ). The solid line shows the best fit of our data with an  $X_3(c)$  law, yielding  $H_{eff} = 190$  kOe. The dashed lines were obtained trying to fit the results with an  $X_2(c)$  law and  $H_{eff} = 100$  kOe, and an  $X_4(c)$  law and  $H_{eff} = 370$  kOe. The insert shows the  $B$  coefficient of the linear term in temperature versus percent cobalt: our results below  $T_0$  (solid circles), Crane's results above  $T_0$  (triangles) (Ref. 3); and corresponding theoretical laws:  $38 N_1 + 520 N_2$  mJ/mole  $K^2$  (solid line) and  $5 + 38 N_1 + 520 N_2$  mJ/mole  $K^2$  (dashed line).

alloys were kept at 77 K and heated to room temperature only for the time necessary to assemble the apparatus. The weighted constituents were such as to produce 0.75-, 1-, 1.5-, 2-, 3-, 4-, and 6-at.% alloys. A very extensive magnetic study (0.04 to 100 K and 0 to 70 kOe) has been made on the same samples and will be published elsewhere.<sup>11</sup> The specific heat was measured with an apparatus using the technique of double-stage demagnetization.<sup>12</sup>

In Fig. 1 we have plotted  $C_p(T)$  curves for five of the seven samples measured. Each curve was observed to fit laws of the type  $C_p = AT^{-2} + BT$ , within experimental error, below the ordering temperature  $T_0$  which was determined from a  $C_p T^2(T^3)$  diagram. The values of  $A$  and  $B$  determined by a least-squares method are plotted as a function of concentration in Fig. 2.

The nuclear term appears to vary almost as the third power of the concentration. This sug-

gests that a magnetic moment may be associated with groups of three or more atoms. More precisely, let  $N_1 = c(1-c)^{12}$ ,  $N_2 = 12c^2(1-c)^{18}$ ,  $N_3 = c^3[24(1-c)^{22} + 36(1-c)^{23} + 90(1-c)^{24}]$  be the probabilities for a Co atom to be isolated or included in groups of two or three atoms, respectively ( $c$  is the concentration); then  $X_2 = c - N_1$ ,  $X_3 = c - (N_1 + N_2)$ ,  $X_4 = c - (N_1 + N_2 + N_3)$  are the concentrations of atoms of Co which are in groups of at least two, three, or four atoms, respectively. Assuming the value of the hyperfine field to be the same for each magnetic cobalt, the variation of the coefficient  $A$  of the  $T^{-2}$  term with the concentration should reflect the variation of the number of magnetic carriers. In Fig. 2 we observe that the best fit of the  $A(c)$  law is obtained with  $X_3(c)$ , so that a magnetic moment is associated with all Co atoms which are included in groups of at least three atoms.

From the expression  $A = [c - (N_1 + N_2)] R[I(I+1)/3] \gamma^2 \hbar^2 H_{\text{eff}}^2 / k^2$ , we calculate for the hyperfine field on the nucleus the value  $H_{\text{eff}} \sim 190$  kOe. This value is not very different from the value of 225 kOe which is observed in hexagonal Co.<sup>13</sup>

But we have neglected the hyperfine specific heat of the matrix. One term is due to the hyperfine field induced by the spin-density oscillations (Ruderman-Kittel-Yosida) of conduction electrons produced by the magnetic cobalt, and so is proportional to  $X_3(c)$ . Assuming this term has the same magnitude for Co as for Fe or Cr impurities in the same matrix,<sup>14</sup> we calculated that the value of  $H_{\text{eff}}$  would be lowered by only 1.5%. Another term (quadrupolar) is due to the electric field gradients associated with the charge-density oscillations (Friedel oscillations) produced by all the Co impurities and is therefore proportional to  $c$ . The fact that we observed a  $c^3$  variation shows that, in our concentration range, this contribution is negligible. However, it may be responsible for some deviation such as is observed for the 0.75 and 1% alloys which give results somewhat higher than the theoretical expectation (Fig. 2). (But the experimental accuracy is poor for the small concentrations and further analysis cannot be made.)

Now we turn to the linear term in temperature  $BT$ , corrected from the contribution of the Au matrix. We assume two different values  $T_1$  and  $T_2$  of the fluctuation temperature of the nonmagnetic impurities depending whether they are isolated or are in pairs. From the spin-fluctuation theory<sup>6,7</sup> they will then contribute<sup>15</sup>  $\gamma_1 T$  (for  $T \ll T_1$ ) and  $\gamma_2 T$  (for  $T \ll T_2$ ) with  $\gamma_1 = N_1(\pi R/T_1)$  and

$\gamma_2 = N_2(\pi R/T_2)$ , respectively, per mole of alloy. The magnetic impurities interacting through long-range oscillations of the  $\cos(2k_F r + \varphi)/r^3$  form will contribute a  $\gamma_3 T$  term which is concentration independent<sup>16</sup> in the range  $T < T_0$  where  $T_0$ , the order temperature, is proportional to their concentration,  $X_3(c)$  in this case.

In the insert of Fig. 2 the values of  $B$  are given for  $T < T_0$  ( $T_0$  is estimated to be around  $8 \times 10^{-2}$  K for the 1.5-at.% alloy from our results). Hence  $B = \gamma_1 + \gamma_2 + \gamma_3$ . On the same graph measurements of  $B$  by Crane<sup>3</sup> at  $T \sim 6$  K, i.e., much greater than  $T_0$ , are also given. In this case  $B = \gamma_1 + \gamma_2$ . Our results differ from Crane's results by an almost constant term of the order of 5 mJ/mole K<sup>2</sup> which is of the right order of magnitude for  $\gamma_3$  (a term of 3.7 mJ/mole K<sup>2</sup> is observed in Cu:Mn and 3 mJ/mole K<sup>2</sup> in Au:Fe<sup>16</sup>).

We can estimate  $\gamma_1$  and  $\gamma_2$  more accurately from Crane's results. He fitted the variation of his  $B$  term with the concentration by the empirical law  $800c^{1.5}$  mJ/mole K<sup>2</sup>. An equally good fit is obtained with  $B = \gamma_1 + \gamma_2 \sim 38N_1 + 520N_2$  mJ/mole K<sup>2</sup> which implies, from what has been said above,  $T_1 \sim 700$  K and  $T_2 \sim 50$  K. It is interesting to note that there exists a logarithmic behavior of the resistivity proportional to  $c$  around 700 K,<sup>17</sup> and that new results<sup>18</sup> show another change of regime below 20 K, where the slope of the increase is proportional to a power 1.5 of the concentration. (Here also this may hide an  $\alpha N_1 + \beta N_2$  variation which may be understood as evidence for short range interactions.)

Thus, our measurements on the Au:Co system confirm the model proposed for the Cu:Co alloys.<sup>4</sup> The agreement is still better: This is not surprising since Au:Co is a better solid solution, so that segregation effects are expected to be of less importance. Our paper supports the suggestion<sup>4</sup> that interaction effects should be of quite general importance in systems near the limiting condition for the appearance of a magnetic moment on the impurities.

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#### QUADRUPOLE MOMENTS OF FIRST EXCITED STATES IN $^{28}\text{Si}$ , $^{32}\text{S}$ , and $^{40}\text{Ar}$ †

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Static quadrupole moments of the first excited states of  $^{28}\text{Si}$ ,  $^{32}\text{S}$ , and  $^{40}\text{Ar}$  have been measured using the reorientation effect in projectile Coulomb excitation. The results obtained are  $Q(^{28}\text{Si}, 2^+) = +0.11 \pm 0.05$  b,  $Q(^{32}\text{S}, 2^+) = -0.20 \pm 0.06$  b, and  $Q(^{40}\text{Ar}, 2^+) = +0.01 \pm 0.04$  b.

The reorientation effect<sup>1</sup> in projectile Coulomb excitation provides a sensitive way to measure quadrupole moments of light nuclei. This method was successfully applied to the measurement of the quadrupole moments of  $^{20}\text{Ne}$  and  $^{22}\text{Ne}$  ( $2^+$  states).<sup>2</sup> In the present work the first excited states in  $^{28}\text{Si}$ ,  $^{32}\text{S}$ , and  $^{40}\text{Ar}$  have been studied using beams from the Berkeley Hilac. These measurements indicate an oblate shape for  $^{28}\text{Si}$ , a prolate shape for  $^{32}\text{S}$ , and a spherical shape, within experimental error, for  $^{40}\text{Ar}$ .

The method used was almost exactly the same as in the Ne experiment.<sup>2</sup> The  $\gamma$ -ray yields from excited projectile and target nuclei at two projectile-scattering angles ( $160^\circ$  and  $90^\circ$ ) were measured simultaneously as particle- $\gamma$  coincidences between a NaI counter ( $7.5 \times 7.5$  cm) and a particle counter at each angle. A multidimensional-analysis program for the Hilac PDP-7 was used to store a  $\gamma$ -ray spectrum, a mixed particle spectrum from the two counters, and a time spectrum. In order to identify the particle counters in off-line analysis, the fast timing signals from one of the particle counters were delayed to pro-

duce two prompt peaks in the time spectra. The ratio of the excitation probability of the projectile to that of the target at each angle was deduced from the measured coincidence  $\gamma$ -ray spectra as  $R^{160} = (N_P^{160}/N_T^{160})(\epsilon_T/\epsilon_P)$ ,  $R^{90} = (N_P^{90}/N_T^{90})(\epsilon_T/\epsilon_P)$ , where  $N$  is the integrated area of a photopeak in the spectra, and  $\epsilon$  is the photopeak efficiency of the NaI counter. Superscripts indicate the scattering angle and subscripts distinguish between projectile ( $P$ ) and target ( $T$ ). A double ratio,  $\mathcal{R} = (R^{160}/R^{90})$ , was also calculated which was less sensitive to many kinds of instrumental effects and to uncertainties in the  $B(E2)$  values of both projectile and target excitations. These ratios, after several corrections were made, were compared with calculated values using the deBoer-Winther Coulomb-excitation program.<sup>3</sup>

This method is advantageous, because the excitation probability of the projectile is considerably more sensitive to its quadrupole moment than that of the target is to its moment. By measuring the target and projectile excitations simultaneously, several kinds of experimental uncer-