CALT-822-93

MAGNETIC FIELD DEPENDENCE OF RESISTIVITY MINIMA IN AMORPHOUS La-Gd-Au ALLOYS WITH HIGH Gd CONTENT

S. J. Poon, J. Durand, and M. Yung

FEBRUARY 1977

A REPORT ON RESEARCH CONDUCTED UNDER CONTRACT FOR THE U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION

W. M. KECK LABORATORY OF ENGINEERING MATERIALS

CALIFORNIA INSTITUTE OF TECHNOLOGY

PASADENA

DISCLAIMER

CALT-822-93

California Institute of Technology

W. M. Keck Laboratory of Engineering Materials

MAGNETIC FIELD DEPENDENCE OF RESISTIVITY MINIMA IN AMORPHOUS La-Gd-Au ALLOYS WITH HIGH Gd CONTENT

by

S. J. Poon, J. Durand and M. Yung

Energy Research and Development Agency Report No. 82, under Contract No. AT(04-3)-822.

Professor Pol Duwez, principal investigator.

FEBRUARY 1977

NOTICE

ABSTRACT

Magnetoresistivity measurements are performed in fields up to 40 kOe on some concentrated amorphous La-Gd-Au alloys exhibiting characteristics of a spin-glass. The negative magnetoresistivity at low temperature is found to be roughly proportional to the square of the magnetization. The resistivity minima in these alloys are attributed to a mechanism of electron scattering from magnetic clouds coupled by the RKKY interactions, in qualitative agreement with analysis of our remanent magnetization data.

Resistivity minima were observed in a large variety of crystalline and amorphous magnetic alloys. Besides the ideal Kondo case of isolated localized magnetic moments diluted in a normal or noble metal[1], such minima occur also in many less-diluted[2] or concentrated[3,4] alloys or compounds[5,6] containing magnetic components. This latter problem has received little attention from theoreticians so far.[7] Meanwhile, resistivity minima in amorphous magnetic alloys have been the subject of many detailed investigations from both experimental and theoretical[8-10] approaches for alloys close to the para-ferromagnetic transition[11,12], and for strongly ferromagnetic alloys as well.[13,14] These minima are such a common phenomenon in amorphous and disordered magnetic alloys that it has been recently suggested that they might be merely structural in origin.[15,16] The apparent insensitivity of the -log T dependent resistivity term to applied fields up to 40 kOe is the only experimental support presented for this latter interpretation. We report on the results of transverse magnetoresistivity measurements on concentrated amorphous $(La_{100-x}Gd_x)_{80}Au_{20}$ alloys ($8 \leq x \leq 40$ at.%) below the onset of long-range ferromagnetic order ($x \simeq 70$). The strong negative magnetoresistivity observed at low temperature leading to the depreciation of the resistivity minima can be correlated to the magnetization data. We conclude that the resistivity minima for our alloys are magnetic in origin.

When Gd is substituted for La in amorphous $(La_{100-x}Gd_x)_{80}Au_{20}$ alloys, a resistivity minimum is observed at $T = T_m$ for $0.5 \leq x \leq 100$. The concentration dependence of T_m was presented in a preliminary report[17] together with the magnetic phase diagram. The "dilute" alloys ($0.3 \leq x \leq 1$) were found to behave like "canonical"

spin-glasses.[18, 19] For higher Gd concentrations up to $x \sim 70$, susceptibility maxima and thermomagnetic history effects were observed. Our remanent magnetization data for the intermediate concentration range ($1 \leq x \leq 40$) were analyzed[20] in terms of magnetic clouds coupled via RKKY interactions.[21] This concentration range, where the magnetization data are still analyzable in a simple way is also most suitable for magnetoresistivity measurements and analysis, for the small positive contribution from the $La_{80}Au_{20}$ matrix becomes negligible as compared with the large negative magnetoresistivity due to Gd. For the dilute region, the normal magnetoresistivity should have been subtracted; and the uncertainties inherent in such a procedure were already pointed out.[22] On the other hand, higher fields are needed to depreciate the resistivity minima in the most concentrated alloys ($x > 40$).

Amorphous samples of nominal composition $(La_{100-x}Gd_x)_{80}Au_{20}$ (with $x = 8$, 12, 16, 20, 34, and 40) were obtained by splat-cooling from the melt. The amorphousness of each foil was checked by X-ray diffraction. Resistivity measurements were performed by using the standard four-probe technique over a temperature range of 2-270°K in zero field, and 2-40°K in transverse fields up to 40 kOe. The resolution of the measurements was 1 part in 10^5. This is sufficient for our purpose since the ratio $(\rho(2^\circ K)-\rho(T_m)/\rho(2^\circ K)$ equals 10^{-3}.

Typical results of the measurements are illustrated in Fig. 1 for $x = 8$ and 40 alloys. General features are observed for the alloys investigated. We comment first on the zero-field data. First, there is no evident correlation between the values of resistivity minima T_m

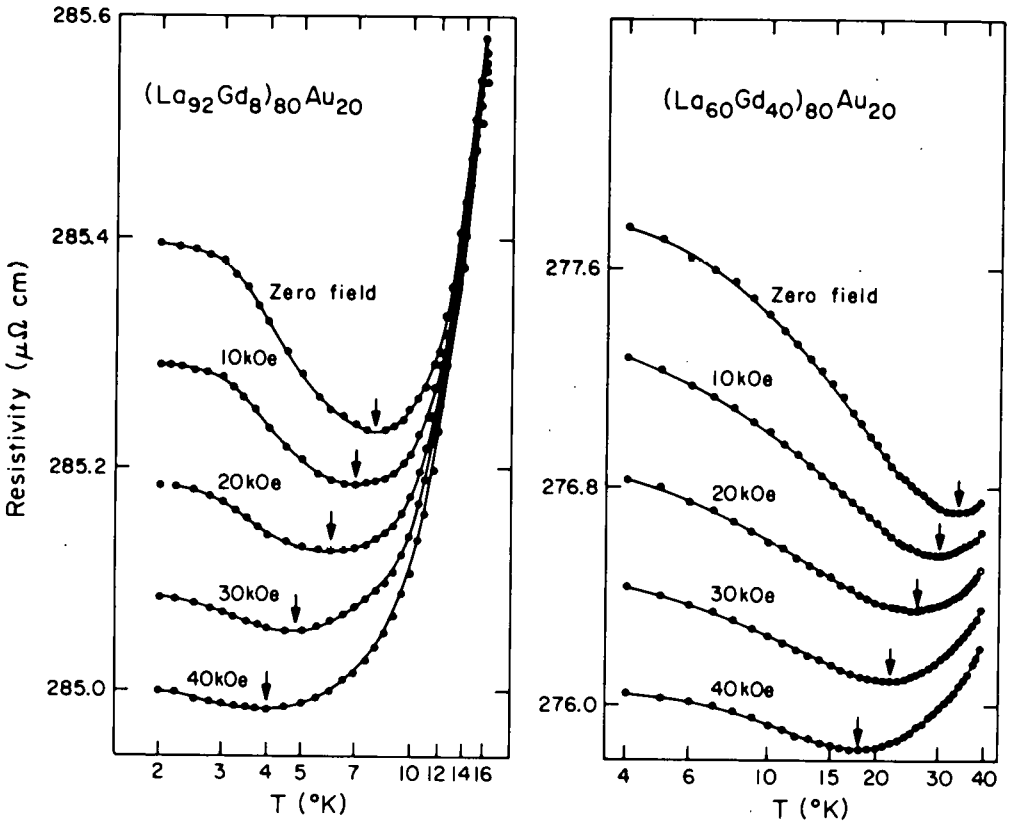

Fig. 1. Resistivity of x = 8 and 40 samples as a function of temperature and magnetic field. Arrows indicate positions of resistivity minima.

at zero field and the temperatures T_M for zero-field susceptibility maxima (Table 1), contrary to the suggestion made in ref. 2. Second, in the vicinity of T_m, the zero-field resistivity may be fitted to a phenomenological equation as:

$$\rho(T) = \rho_o - A \log T + BT^2 \qquad (1)$$

At lowest temperatures, one observes a departure from logarithmic dependence and a tendency towards saturation. A rough estimate of the anomalous part of the zero-field resistivity is given by $\rho(2^oK)-\rho(T_m)$; this quantity increases noticeably with Gd concentration (Table 1).

In magnetic fields up to 40 kOe, the coefficient B in equation (1) remains practically constant within the limits of experimental accuracy, so that the T^2 contribution does not originate from localized spin-fluctuations scattering[23] nor from electron-magnon collisions with spin-flips.[24] It is more likely to arise from either phonon effects[8] or electron-electron Baber scatterings[25] as to be investigated. In contrast, the low-temperature anamolous resistivity is drastically suppressed by an applied field. Although no resistivity maxima are observed in high fields as in Cu-Fe, CuMn dilute alloys[26], yet our results are strongly reminiscent of those observed in other classical Kondo systems like Cu-Cr[27], Cu-Au-Fe, Cu-Au-Cr.[28] This analogy motivates us to analyze our magnetoresistivity measurements in conjunction with the magnetization data following the procedure used for dilute Kondo alloys. The justification follows in the discussion. Such an approach was previously found to be rather successful in some other concentrated amorphous alloys.[11]

TABLE 1. Resistivity and magnetization data for amorphous concentrated $(La_{100-x}Gd_x)_{80}Au_{20}$ alloys.

Concentration (x at.% Gd.)	T_M (^oK)	T_m (^oK)	$\rho(2^oK)-\rho(T_m)$ ($\mu\Omega$ cm)	$\rho(H = 0)-\rho(40$ kOe) at 2.0^oK ($\mu\Omega$cm)	μ^* (μ_B)	C^* (at.%)	θ_c (^oK)	$\overline{J}_{sf}$ (eV)
8	3.2	8	0.15	0.38	12	4.7	5.0	0.09
12	4.8	7	0.26	1.45	15	5.5	9.5	0.13
40	21.5	34	~ 1.25	~ 1.80	42	7.0	36	0.06

As shown in Fig. 2 for a sample of $x = 40$, the magnetoresistivity data $\Delta\rho_m = \rho(H,T) - \rho(0,T)$ at low temperatures ($T \leq 16^{\circ}K$, for $x = 40$ alloy) may be fitted to a unique function of $H/(T + \theta_c)$, the parameter θ_c being concentration dependent. Similarly, the magnetization $M(H,T)$ ($H_{max} = 70$ kOe) follows a unique function of $H/(T + \theta_c)$ over the low-temperature range ($T \leq 25^{\circ}K$ for $x = 40$ alloy). This function is a Brillouin function with $\mu^* = gJ\mu_B = 42\mu_B$ (practically identical to a Langevin function in this case) for a $x = 40$ sample (Fig. 3). Values of θ_c and μ^* for $x = 8$, 12, and 40 are listed in Table 1. Such fittings fail at higher temperatures which probably indicates dissociation of μ^* into smaller moments. Finally, $\Delta\rho_m$ was found to be roughly proportional to M^2 (Fig. 4), so the classical relations derived for the dilute Kondo alloys are fairly well verified in our concentrated amorphous alloys for $(g\mu_B H/(T + \theta_c)) \leq 2$:

$$|\Delta\rho_m| \sim M^2 \sim [B_J(\mu^* H/k(T + \theta_c))]^2 \qquad (2)$$

The apparent linearity of $\Delta\rho_m$ as a function of $H/(T + \theta_c)$ in Fig. 2 originates from the fact that the Brillouin function is rather parabolic at intermediate values of $H/(T + \theta_c)$ as can be seen in Fig. 3. The proportionality between $\Delta\rho_m$ and M^2 has been observed long time ago.[29] This relation was found to be valid in concentrated magnetic alloys[30, 11, 12] as well as in the dilute cases.[26-28] In the case of non-interacting moment, this result is rather general for any scattering process which involves spin flips of the conduction electrons and impurity spins.[31, 32] The coefficient of proportionality between $\Delta\rho_m$ and M^2 is derived in the limit of $(g\mu_B H/kT) \ll 2$, or $(g\mu_B H/(T + \theta_c)) < 2$

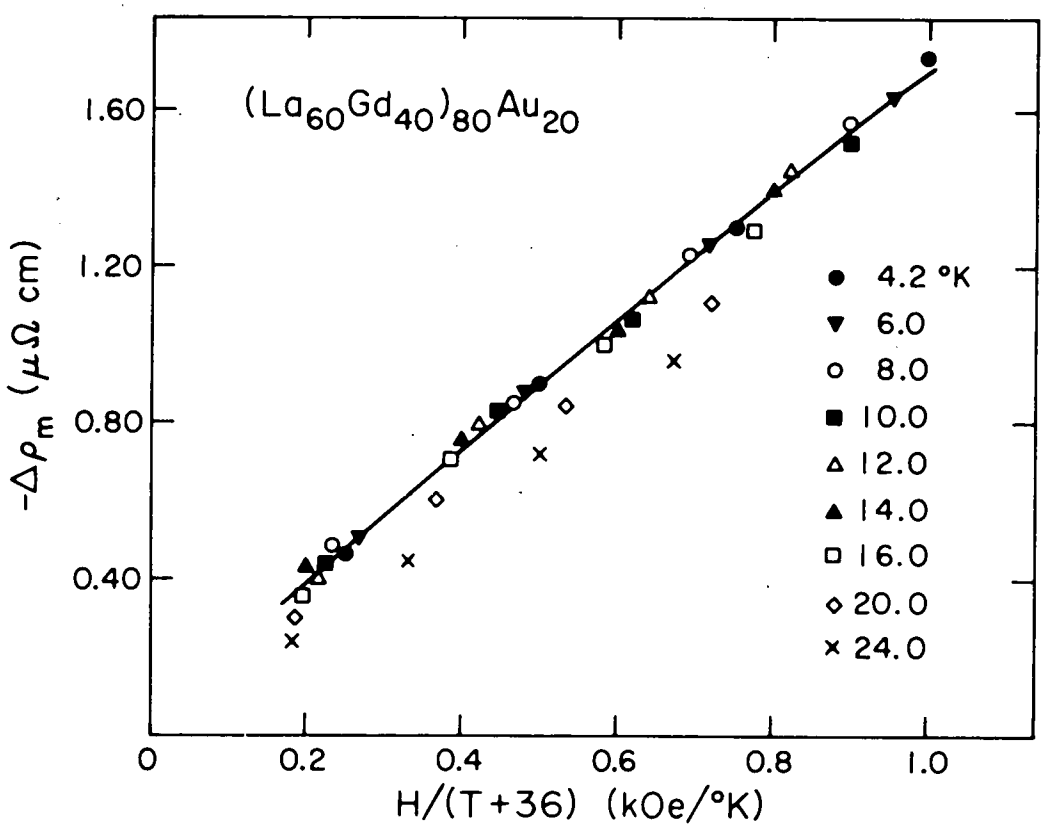

Fig. 2. Negative magnetoresistivity of the 40 at.% sample vs H/(T + 36). Solid line indicates the general behavior below 16°K.

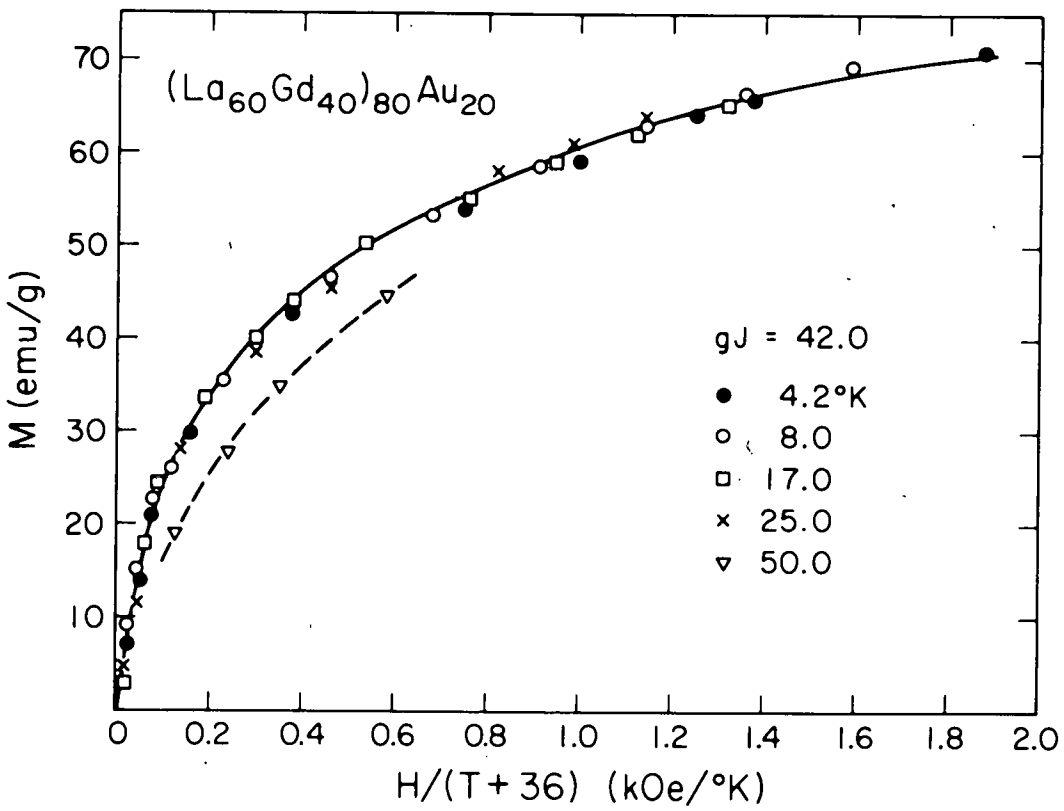

Fig. 3. Magnetization of the 40 at.% sample vs H/(T + 36. Solid line is the Brillouin function fit with g = 2, J = 21, and θ_c = 36°K. Dashed line shows deviation from such relation.

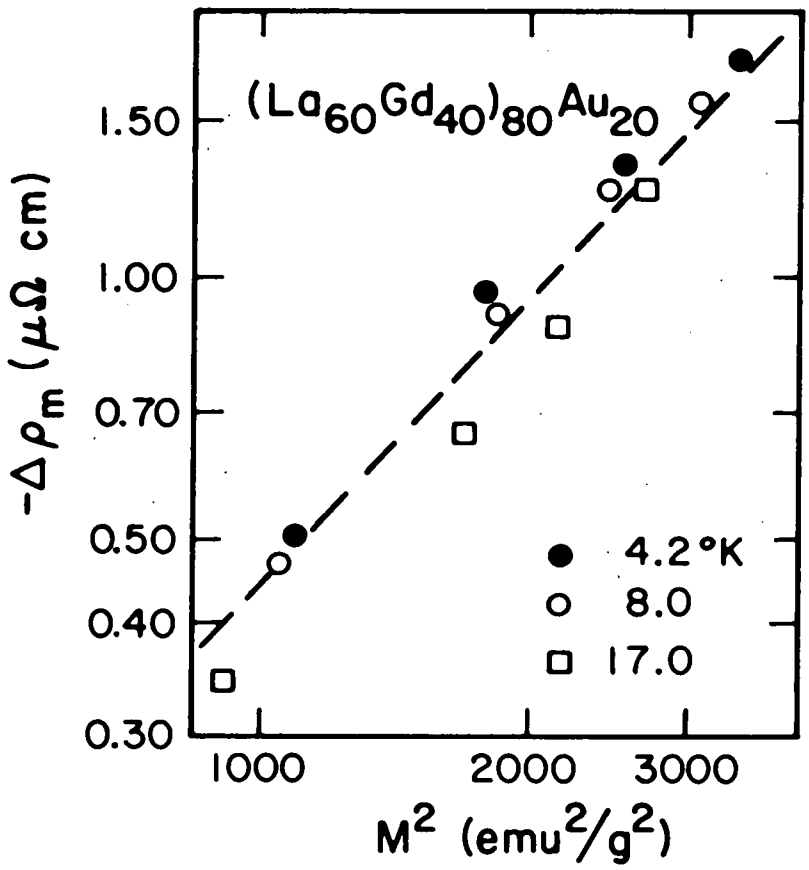

Fig. 4. Negative magnetoresistivity of the 40 at.% sample vs its magnetization squared. Dashed line is an arbitrary line of slope unity.

if one replaces T by an effective thermodynamic temperature $T + \theta_c$ and in first order perturbation as:

$$|\Delta\rho_m| = (3\pi m/2E_F e^2\hbar)\, c\, V_o J_{sf}^2 M^2 [1 + (\mu_B/\mu_{eff})^2] \qquad (3)$$

where the notations follow those in ref. 32. The fact that the averaged magnetic clouds μ^* obey a Brillouin function in equation (2) indicate that they may be regarded as pseudo single magnetic entities at the effective temperature $T + \theta_c$. It is tempting to apply the Beal-Monod-Weiner results (equation 3) to the case of non-interacting magnetic clouds by replacing c by c*, the atomic concentration of the clouds (Table 1); μ_{eff} by μ^*, the size of the clouds; V_o by V_o^*, the "atomic" volume of the clouds. By doing so, we find that the values obtained for $\overline{J}_{sf}$, the exchange constant between the conduction electrons and the magnetic clouds, ($0.06 < \overline{J}_{sf} < 0.13$ eV, see Table 1) agree reasonably well with the values of J_{sf} as determined for dilute La-Gd-Au alloys from the approach to saturation magnetization ($J_{sf} = 0.14$ eV) and from the initial depression of the superconducting transition ($J_{sf} = 0.16$ eV).[18] We use $E_F \sim 7$ eV as in ref. 18.

A detail comparison between the present analysis and that for the remanent magnetization[20] on the same samples is not feasible at this moment. The scaling laws hold for the remanent magnetization while they do not for high field magnetization and consequently for high field magnetoresistivity over this concentration range. From the analysis of the remanent magnetization data, we were able to describe our samples at low fields as an assembly of independent magnetic clouds, whose average size was concentration independent and contains $\sim$ 50

spins. However, the uncompensated moment (net moment) in each cloud is equivalent to $\sim$ 7 spins. The high field measurements give a picture of magnetic clouds considerably smaller in size and concentration dependent (see Table 1). It is possible that in high fields, magnetic interactions on a finer scale are manifested. Despite these differences, it is interesting to note that the magnetic properties of these spin-glass alloys in both the low-field and high-field limits may be explained by the same phenomenological approach of magnetic clouds indirectly coupled via the RKKY interaction.[21] As mentioned by many authors, the fit of magnetization data to a modified Brillouin function with an effective temperature $T + \theta_c$ is lacking in theoretical justification. This parameter θ_c determined at higher fields cannot be correlated in a priori with any ordering temperature. However, the concentration dependence of this parameter θ_c is characteristic of the spin-glass regime, since in Kondo alloys such a θ_c is practically concentration independent and gives the order of magnitude of T_K.[21]

Let us emphasize also that the use of a single-impurity formalism for our analysis in terms of scattering from magnetic clouds does not allow us to draw any hasty conclusion about the single-impurity state of Gd in the $La_{80}Au_{20}$ matrix. For example, Ni does not carry any moment in the Hartree-Fork sense when diluted in Cu or V. On the other hand, Fe has a localized spin-fluctuation behavior[33] when diluted in Rh. But the concentrated Ni-Cu[3], Ni-V[4], Rh-Fe[2] alloys exhibit resistivity minima which may be explained by the common presence in these alloys of magnetic entities (giant moments or magnetic clouds) having a Kondo-like behavior. In the case of Gd in $La_{80}Au_{20}$, high field magnetization measurements showed that for concentration as

low as 0.24 at.% the Gd has its full moment of 7 μ_B without any magnetic clustering effect. From electronic structure point of view[36], a Kondo state for single Gd is believed to be unlikely, contrary to the case of rare-earth with almost empty or almost filled f shells[35] (Ce, Yb). However, let us note that the giant moments systems are usually described by a ferromagnetic exchange coupling between the spin S of the impurity and the spin s of the conduction electron. Such a Hamiltonian overlooks the possible existence of a Kondo-like singlet state which would imply an antiferromagnetic coupling at low temperature. In fact, recent nuclear orientation experiments yielded evidence for a Kondo-like state for giant moments in Pt-Co and Pd-Co alloys.[36] Finally, let us mention that the resistivity behavior of "less-diluted" alloys was satisfactorily explained by calculations of the electron scattering by pairs of magnetic atoms.[37] As it has been already suggested[3-5, 11, 12] that such an approach extended to larger magnetic entities may lead to an understanding of resistivity minima in concentrated magnetic alloys in the amorphous as well as in the crystalline state.

ACKNOWLEDGEMENT - The authors wish to thank Professor Pol Duwez for his support throughout this work.

REFERENCES

*Work supported by the Energy Research Development Agency, Contract No. AT(04-3)-822.

†On leave from Laboratoire de Structure Electronique des Solides, E.R.A. 100, 4, rue Blaise Pascal, 67000 Strasbourg, France.

1. KONDO, J., Solid State Phys. 23, 183 (1969); HEEGER, A. J., id. 283 (1969); for most recent reviews, see Magnetism V, Rado and Suhl, edit., (Academic Press, New York, 1973); also, ORBACH, R,, AIP Conf. Proc. 24, 3 (1974).

2. MURANI, A. P. and COLES, B. R., J. Phys. C: Metal Phys. Suppl. 2, S159 (1970) (Rh-Fe); SARKISSIAN, B. V. B. and TAYLOR, R. H., J. Phys. F 4, L243 (1974) (Pt-Mn).

3. HOUGHTON, R. W., SARACHIK, M. P. and KOUVEL, J. S., Solid State Commun. 8, 943 (1970) (Ni-Cu); CLAUS, H. and MYDOSH, J. A., Solid State Commun. 14, 209 (1974) (Fe-V), SMITH, T. F., TAINSH, R. J., SHELTON, R. N. and GARDNER, W. E., J. Phys. F 5, L96 (1975) (Ni-Cr).

4. LABORDE, O., LOEGEL, B. and RADHAKRISHNA, P., J. Physique (Paris) 35, C4-247 (1974) (Pt-Co, Ni-V).

5. YAMAGUCHI, Y., AOKI, T. and BRITTAIN, J. O., J. Phys. Chem. Solids 31, 1325 (1970); IKEDA, K., Phys. Lett. 53A, 375 (1975) (NiAl).

6. SELLMYER, D. J., CASKEY, G. R. and FRANZ, J., J. Phys. Chem. Solids 33, 561 (1972) (CoAl); CASKEY, G. R., FRANZ, J. M. and SELLMYER, D. J., J. Phys. Chem. Solids 34, 1179 (1973) (FeAl, NiAl); AMAMOU, A., Thesis, Strasbourg (1975) (CoSi).

7. RIVIER, N., J. Phys. F4, L249 (1974); KIM, D. J. and SCHWARTZ, B. J., AIP Conf. Proc. 24, 456 (1974) and Phys. Rev. B (1977, to be published).

8. HASEGAWA, R., Phys. Lett. 36A, 207 (1971) also 36A, 425 (1971); MADHUKAR, A. and HASEGAWA, R., J. Phys. (Paris) 35, C4-291 (1974).

9. KANEYOSHI, T. and HONMURA, R., Phys. Lett. 46A, 1 (1973).

10. SILVER, R. N. and MCGILL, T. C., Phys. Rev. B9, 272 (1974).

11. TSUEI, C.C. and HASEGAWA, R., Solid State Commun. 7, 1581 (1969); HASEGAWA, R. and TSUEI, C. C., Phys. Rev. B2, 1631 (1970); 3, 214 (1971).

12. LIANG, V. K. C. and TSUEI, C. C., Solid State Commun 9, 579 (1971); Phys. Rev. B7, 3215 (1973).

13. LIN, S. C. H., J. Appl. Phys. 40, 2173 (1969); HASEGAWA, R. and DERMON, J., Phys. Lett. 42A, 407 (1973); YOSHIIE, T., YAMAKAWA, K. and JUJITA, F. E., J. Phys. Soc. Japan 37, 572 (1974); LOGAN, J. and YUNG, M. J. Non-Cryst. Solids 21, 151 (1976); see also papers in Amorphous Magnetism II, Troy, New York, 1976 (Plenum Press, New York, 1977, to be published).

14. RAPP, O., BHAGAT, S. M. and JOHANNESSON, C., Solid State Commun. 21, 83 (1977).

15. COCHRANE, R. W., HARRIS, R., STRÖM-OLSEN, J. O. and ZUCKERMANN, M. J., Phys. Rev. Lett. 35, 676 (1975). Recent acoustic experiments did not support this model, see BELLESSA, G., J. Phys. (Paris) 37, L-291 (1976).

16. KONDO, J., Physica 84B, 40 and 207 (1976).

17. POON, S. J. and DURAND, J., in Amorphous Magnetism II, Troy, New York, 1976 (Plenum Press, New York, 1977, to be published.

18. POON, S. J. and DURAND, J., Solid State Commun. 21 (1977, to be published). "Canonical" in the sense that certain thermodynamically derived quantities (e.g. magnetization, specific heat, susceptibility,....) obey the scaling laws.

19. SOULETIE, J. and TOURNIER, R., J. Low-Temp. Phys. 1, 95 (1969).

20. POON, S. J. and DURAND, J., Commun, Phys. 2 (1977, to be published).

21. THOLENCE, J. L., and TOURNIER, R., J. Phys. (Paris) 35, C4-229 (1974) and in ref. 17; HOLTZBERG, F., THOLENCE, J. L., and TOURNIER, R., in Int. Conf. Mag., Amsterdam, 1976 (to be published).

22. ALDERSON, J. E. A. and HURD, C. M., J. phys. Chem Solids 32, 2075 (1971); HURD, C. M., and ALDERSON, J. E. A., Phys. Rev. B4, 1088 (1971) and B6, 1894 (1972).

23. KAISER, A. B. and DONIACH, S., Intern. J. Magnetism 1, 11 (1970); PURWINS, H. G., SHULZ, H., and SIERRO, J., Intern. J. Magnetism 2, 153 (1972).

24. GOODINGS, D. A., Phys. Rev. 132, 542 (1963).

25. BABER, W. G., Proc. Phys. Soc. A, 158, 383 (1937).

26. MONOD, P., Phys. Rev. Lett. 19, 1113 (1967).

27. DAYBELL, M. D. and STEYERT, W. A., Phys. Rev. Lett. 20, 195 (1968).

28. FENTON, E. W., Phys. Rev. B5, 3788 (1972); B7, 3144 (1973); B10, 4805 (1974).

29. KORRINGA, J. and GERRITSEN, A. N., Physica 19, 457 (1953).

30. SCHMITT, R. W. and JACOBS, I. S., J. Phys. Chem. Solids 3, 324 (1957).

31. YOSHIDA, K., Phys. Rev. 107, 396 (1957).

32. BEAL-MONOD, M. T. and WEINER, R. A., Phys. Rev. 170, 552 (1968); B3, 145 (1971); NAGAI, S. and KONDO, J., J. Phys. Chem. Japan 38, 129 (1975).

33. RUSBY, R. L., J. Phys. F4, 1265 (1974).

34. BLANDIN, A., in Magnetism V, Rado and Suhl, edit., (Academic Press, New York, 1973) p.76.

35. CORNUT, B. and COQBLIN, B., Phys. Rev. B5, 4541 (1972); BHATTACHARJEE, A. K. and COQBLIN, B., Phys. Rev. B13 3441 (1976); SAMWER, K. and WINZER, K., Z. Physik B25, 269 (1976), and references therein.

36. FLOUQUET et al., Phys. Rev. Lett. 38, 81 (1977) and references therein.

37. BEAL-MONOD, M. T., Phys. Rev. 178, 874 (1969); MATHO, K., and BEAL-MONOD, M. T., Phys. Rev. B5, 1899 (1972).