order, no evidence that points to an abrupt change in electronic structure at the metal-insulator transition. In fact the conduction processes appear to fall within the description of percolation theory. In a broader context the present results cast grave doubts on theories currently employed to describe the coupling between local optical excitations and the conduction electrons of a metallic host. This coupling, whose character near E_F assumes a pivotal role in determining the *theoretical* threshold profile,[11] appears much less important in the present *experimental* studies. Whereas translational invariance and metallic properties of the host are central to the theoretical models, it appears in practice that the local short-range ordering of atoms has the major influence on the optical excitation spectra of real alloys. Future theories that reflect these differing emphases will provide a more fundamental understanding of optical processes in conducting solids.

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[1]Discussions of this area are collected in Proceedings of the International Conference on the Metal-Nonmetal Transition, San Francisco, California, 1968, Rev. Mod. Phys. 40, 673–844 (1968).

[2]R. C. Cate, J. G. Wright, and N. E. Cusack, Phys. Lett. 32A, 467 (1970); H. Endo, A. I. Eatah, J. G. Wright, and N. E. Cusack, J. Phys. Soc. Jpn. 34, 666 (1973).

[3]K.-F. Berggren and G. Lindell, Solid State Commun. 13, 1589 (1973).

[4]B. Raz, A. Gedanken, U. Even, and J. Jortner, Phys. Rev. Lett. 28, 1643 (1972).

[5]R. A. Tilton and C. P. Flynn, preceding Letter [Phys. Rev. Lett. 34, 20 (1975)].

[6]R. A. Tilton, D. J. Phelps, and C. P. Flynn, Phys. Rev. Lett. 32, 1006 (1974).

[7]H. Wetzl, in *Vacancies and Interstitials in Metals*, edited by A. Seeger *et al.* (North-Holland, Amsterdam, 1970).

[8]H. Scher and R. Zallen, J. Chem. Phys. 53, 3959 (1970).

[9]S. Kirkpatrick, Phys. Rev. Lett. 27, 1722 (1971), and in *Theory of the Properties of Metals and Alloys*, edited by S. Takenchi (Halsted Press, New York, 1973).

[10]G. Baldini, Phys. Rev. 128, 1562 (1962).

[11]For an extensive discussion see G. H. Mahan, in "Solid State Physics" (Academic, New York, to be published), Vol. 29.

Ferromagnetic Transition in V-Fe

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Low-field magnetization measurements on V-Fe alloys (28.5 and 29.5 at.% Fe) reveal a complex magnetic behavior of these alloys. However, the reversible part of the paramagnetic susceptibility indicates a well-defined ferromagnetic transition. Below the transition temperature, T_c, activation processes at very low fields cause the initial susceptibility to decrease rapidly as the temperature is lowered below T_c, thus explaining why the ac susceptibility displays a sharp peak at T_c.

The onset of long-range ferromagnetism in disordered V-Fe alloys is attributed to giant spin clusters.[1,2] When Fe is added in dilute amounts to V no localized moments are observed.[1,2] With increasing Fe concentration, because of statistical reasons, more and more Fe atoms are located in highly Fe-rich atomic environments. It is assumed that these Fe-rich regions can polarize spontaneously, forming giant spin clusters.[1,2] It is the interaction between these giant clusters which can lead to the onset of long-range ferromagnetism at higher Fe concentration.

There are several alloy systems in which magnetic properties are presently understood in terms of giant spin clusters, the most extensively studied being Ni-Cu.[3] Like all of these alloys, V-Fe exhibits large specific-heat anomalies for concentrations near the onset of magnetic order.[4-6] It is assumed that these anomalies are due to a local anisotropy of the giant moments, often approximated by an effective magnetic field, giving rise to a splitting of the Zeeman levels of the giant spin moments.[3,5-7]

The exponential decrease of the specific-heat

anomaly of V-Fe alloys below 1°K indicates a level splitting of the order of $\Delta E/k_B = 1$°K.[8] Since the spin clusters are assumed to have a moment of about $20\mu_B$,[2] one would expect that the anisotropy energy, i.e., the energy difference between the easy and hard directions of the cluster moment, is at least several times the level splitting, or several degrees.

If the exchange interaction between the spin clusters is of the same order as or even smaller than the local anisotropy, the onset of long-range order may be greatly affected or even prevented. Any distribution of cluster size and interaction strength could further complicate the magnetic transition.

Recently, it has been shown that V-Fe alloys with a concentration near 27 at.% Fe display a sharp peak in the ac susceptibility.[9] Similar peaks have been found in several spin-glass or mictomagnetic alloys[10,11] and are assumed to signal the onset of a random freezing of the spin moments.

To investigate the nature of the magnetic order in V-Fe alloys, I performed low-field magnetization measurements on two V-Fe alloys with concentrations of 28.5 and 29.5 at.% Fe. The temperature of the onset of magnetic order has been reported to be about 4 and 16°K, respectively,[9] and is therefore in a convenient temperature range for detailed magnetization measurements. The alloys have been arc-melted, annealed for four days at 1200°C in a sealed quartz capsule, and water-quenched. Since the magnetic properties are very sensitive to small changes in the Fe concentration,[12] and also to changes in the metallurgical state (chemical short-range order),[1,13] great care had to be taken with the preparation of the samples. The sharpness of the ac susceptibility peak[9] is probably the most sensitive indication of a homogeneous alloy and the samples used for the dc magnetization measurement were selected accordingly. The two samples for the measurement were taken from earlier resistivity samples[9] and were small rectangular cylinders of 6 and 15 mg weight with a length-to-diameter ratio of about 3 and 5, respectively.

The magnetization measurements were performed with a vibrating-sample magnetometer with a continuous-flow cryostat. The absolute accuracy for the magnetization measurements is $\pm 10^{-5}$ emu and for the temperature ± 0.05°K. Measurements of the ac susceptibility were performed by an induction method at a frequency of 38 Hz and a field amplitude of 0.1 Oe.

The results shown in Fig. 1 demonstrate the complex magnetic behavior of the V-Fe alloys. The upper part of Fig. 1 represents the magnetization at a constant field of 3 Oe as a function of temperature for both decreasing and increasing temperature, the latter being measured after initially cooling the sample in zero field from 50 to 4.2°K. Also shown for comparison is the ac susceptibility in arbitrary units. The large difference in the magnetization between field-cooled and zero-field-cooled states (thermomagnetic history effect) is similar to that previously ob-

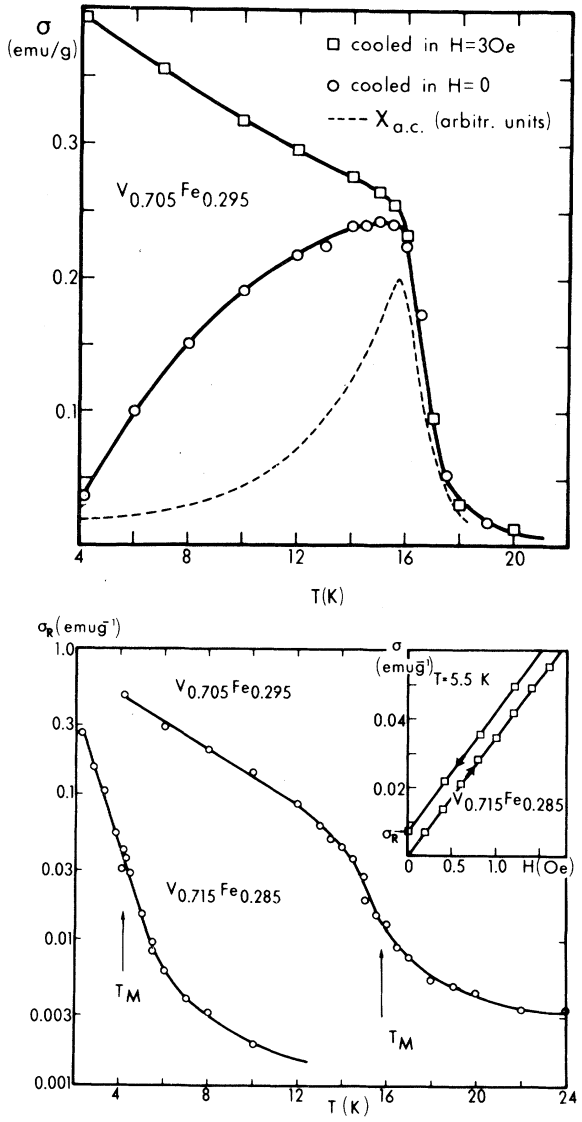

FIG. 1. Upper part, magnetization at constant field: □, for decreasing temperatures; ○, for increasing temperatures; ----, ac susceptibility. Lower part, isothermal remanence versus temperature; inset, magnetic isotherm for increasing and decreasing field.

served in several alloy systems where it has been been discussed in terms of a mictomagnetic or spin-glass state.[14-16]

Irreversible processes are observed in both V-Fe alloys up to high temperatures. The lower part of Fig. 1 shows the temperature dependence of the isothermal remanence σ_R in the form of a semilogarithmic plot. σ_R was measured after initially cooling the sample in zero field to each temperature, applying isothermally a field of 200 Oe, and removing it again. Indicated by arrows are the temperatures T_M where the two alloys show a sharp peak in the ac susceptibility. The remanent magnetization certainly does not indicate that the alloys undergo any magnetic phase transition at T_M. At all temperatures the remanent magnetization depends explicitly on time, decreasing about 10% over the period of 1 h, indicating very slow relaxation processes.[15-17] The insert in Fig. 1 shows a magnetization curve typical for a temperature above T_M. The curve was obtained after cooling the sample to 5.5°K in zero field (from 50°K). The initial linear increase of the magnetization is reversible up to about 2 Oe. At higher fields irreversible processes set in. The field in this example was increased to 200 Oe and then lowered to zero. As can be seen in the inset of Fig. 1, the curves for increasing and decreasing field are parallel, displaced by the isothermal remanence σ_R, which, in turn, very slowly decays. This behavior suggests that the true equilibrium susceptibility can best be determined from the initial slope of the magnetization curves taken with increasing field after cooling first in zero field to the temperature in question. It also demonstrates that measurement of the magnetization taken at constant field as a function of temperature, as that shown in the upper part of Fig. 1, may not be very definitive in studying equilibrium properties.

Figure 2 shows the results of the magnetization measurement taken with increasing field (starting from the zero-field–cooled state). The upper part of Fig. 2 shows a typical set of isotherms for one of the alloys. The magnetization is shown as function of the applied field, H_a. Starting from the highest temperature of 18°K (see inset in Fig. 2), σ versus H_a rises linearly from the origin with a slope (initial susceptibility) which rapidly increases with decreasing temperature, reaching a sudden limiting value at 15.8°K. During this fast increase in the slope, the nature of the magnetization process near $H_a = 0$ is gradually changing. Irreversible processes are setting in, giving rise to a time-dependent magnetization and causing some curvature in σ versus H_a near $H_a = 0$; i.e., the linear part of the σ-versus-H_a curves no longer extrapolate back to the origin but intercept the H_a axis at a finite field (a coercive force). This coercive force increases very fast as the temperature is lowered further (Fig. 2), causing the magnetization σ to increase very slowly with small fields. The coercive force does not suddenly set in as the temperature is lowered, but rather develops very gradually (at 16.3°K it is just barely outside the experimental accuracy).

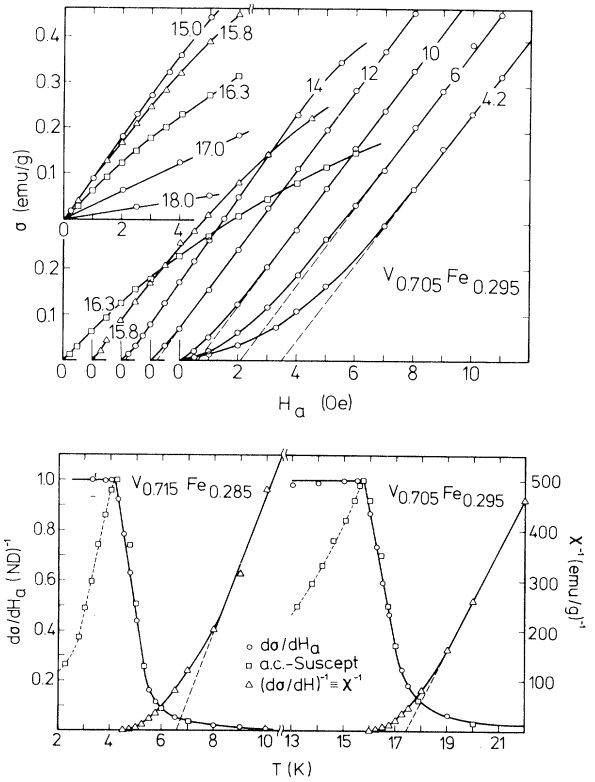

FIG. 2. Upper part, magnetic isotherms versus applied field; numbers indicate temperatures (°K). Lower part, results of the magnetic isotherms; $d\sigma/dH_a$ represents the maximum slope of each isotherm.

These results demonstrate the increasing importance of the anisotropy on the low-field magnetization as the temperature is lowered. The variation of $d\sigma/dH_a$ near $H_a = 0$ with temperature explains why the ac susceptibility displays a peak near 16°K. The detailed shape of the ac susceptibility near the peak is probably determined by nonequilibrium properties, since near 16°K, as mentioned above, irreversible processes in the magnetic isotherms set in at very low fields.

The maximum value of the initial susceptibility which was reached near 16°K limits the rate of increase of all isotherms at lower temperature (the straight-line portions of all isotherms below 16°K are all parallel). This limiting value of $d\sigma/dH_a$ is determined by the geometry of the sample and is equal to $1/ND$ (N is the demagnetization factor, and D is the density). The magnetization is determined by the magnetic field H inside the sample. Since $H = H_a - ND\sigma$,

$$\frac{d\sigma}{dH_a} = \frac{d\sigma}{dH}\frac{dH}{dH_a} = \frac{d\sigma}{dH}\left(1 - ND\frac{d\sigma}{dH_a}\right); \quad (1)$$

solving for $d\sigma/dH_a$ and setting $d\sigma/dH = \chi$ yields

$$d\sigma/dH_a = \chi/(1+ND\chi) \leq 1/ND. \quad (2)$$

The maximum value $(ND)^{-1}$ of $d\sigma/dH_a$ is reached when the susceptibility χ diverges. This limiting value for $d\sigma/dH_a$, determined from the slope of the straight-line sections of the isotherm at low temperatures, is 0.214 and 0.094 emu/g for the alloys with 28.5 and 29.5 at.% Fe, respectively.

The lower part of Fig. 2 summarizes the results of the magnetization measurements for both alloys. Shown is $d\sigma/dH_a$, in units of $(ND)^{-1}$, determined at high temperatures from the initial slope of the magnetic isotherms and at low temperatures from the straight-line sections of the isotherms. As the temperature is lowered, both alloys reach this limiting value of $d\sigma/dH_a$ rather abruptly. This behavior is usually assumed to be a good indication of a well-defined ferromagnetic phase transition[18] (the transition is sharp within the experimental accuracy of 0.05°K, giving $T_c = 4.2$ and 16.8°K for the 28.5- and 29.5-at.%-Fe samples, respectively). Also shown in Fig. 2 is the normalized ac susceptibility. The temperature of its maximum, T_M, agrees well with T_c, the temperature of the kink in $d\sigma/dH_a$. The paramagnetic initial susceptibility (above T_c) has been corrected for demagnetization effects [by using Eq. (2)] and is also shown in Fig. 2 in reciprocal form ($1/\chi$ versus T, scale on the right ordinate). The strong curvature near T_c reflects the complicated nature of the magnetic short-range order above T_c.[19]

In conclusion I have shown that V-Fe alloys, which exhibit spin clusters with complicated magnetic interactions, undergo a well-defined ferromagnetic transition, but that this transition can be masked by nonequilibrium processes with long relaxation times. Furthermore, I have shown that the observed sharp peak in the ac susceptibility of these alloys is due to activation processes at very low fields. These processes set in right at the ordering temperature, T_c, causing the initial susceptibility to decrease rapidly as the temperature is lowered below T_c.

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[1]W. J. Nellis and M. B. Brodsky, J. Appl. Phys. 43, 1961 (1972).

[2]P. Pataud and R. Tournier, to be published.

[3]For a recent discussion of all the relevant experiments in Ni-Cu alloys see, for example, K. Levin and D. L. Mills, Phys. Rev. B 9, 2354 (1974).

[4]C. H. Cheng, C. T. Wei, and P. A. Beck, Phys. Rev. 120, 426 (1960).

[5]A. Hahn and E. P. Wohlfarth, Helv. Phys. Acta 41, 857 (1968).

[6]J. S. Kouvel, *Magnetism in Alloys* (Metallurgical Society of the American Institute of Mining, Metallurgical, and Petroleum Engineers, New York, 1972).

[7]K. Schröder, J. Appl. Phys. 32, 880 (1961); J. D. Livingston and C. P. Bean, J. Appl. Phys. 32, 1964 (1961).

[8]R. G. Scurlock and E. M. Wray, Phys. Lett. 6, 28 (1963); W. Proctor and R. G. Scurlock, in *Proceedings of the Eleventh Conference on Low Temperature Physics, St. Andrews, Scotland, 1969,* edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews Univ. Press, St. Andrews, Scotland, 1969), p. 1320.

[9]H. Claus and J. A. Mydosh, Solid State Commun. 14, 209 (1974).

[10]V. Cannella, J. A. Mydosh, and J. I. Budnick, J. Appl. Phys. 42, 1689 (1971); V. Cannella, in *Amorphous Magnetism,* edited by H. O. Hooper and A. M. de Graff (Plenum, New York, 1973), p. 195.

[11]A. Mukhopadhyay and P. A. Beck, to be published.

[12]A change of the Fe concentration from 28.5 to 29.5 at.% Fe changes T_M, the temperature of the susceptibility maximum, by a factor of 4 (from about 4 to 16 °K); see Ref. 9.

[13]R. S. Preston, D. J. Lam, M. V. Nevitt, D. O. van Ostenburg, and C. W. Kimball, Phys. Rev. 149, 440 (1966).

[14]P. A. Beck, *Magnetism in Alloys* (Metallurgical Society of the American Institute of Mining, Metallurgical, and Petroleum Engineers, New York, 1972), p. 211.

[15]A. P. Murani, J. Phys. F: Metal Phys. 4, 757 (1974).

[16]J. L. Tholence, Ph.D. thesis, Paris, 1973 (unpublished).

[17]P. A. Beck, J. Less-Common Metals 28, 193 (1972).

[18]A. Arrott and J. E. Noakes, in *Iron and Its Dilute Solid Solutions,* edited by C. W. Spencer and F. E. Werner (Interscience, New York, 1963), p. 8; S. Arajs and R. V. Colvin, J. Appl. Phys. 35, 2424 (1964).

[19]A large value of γ has been recently reported for the giant moment system NiRh (W. C. Muellner and J. S. Kouvel, to be published) and for PdFe [J. S. Kouvel and J. B. Comly, *Critical Phenomena in Alloys, Magnets, and Superconductors,* edited by R. E. Mills, E. Ascher, and R. I. Jaffee (McGraw-Hill, New York, 1971), p. 437].