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HIGH FIELD MAGNETISM

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Osaka, Japan, September 13-14, 1982

Edited by

M. DATE

High Magnetic Field Laboratory

Faculty of Science

Osaka University

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HIGH FIELD MAGNETISM

PREFACE

The International Symposium on High Field Magnetism was held at the Osaka University and Hotel Plaza in Osaka, September 13-14, 1982 as a satellite symposium of the International Conference on Magnetism-1982-Kyoto. More than 100 participants, including high international representation attended the two-day Symposium which consisted of one poster session and seven plenary sessions including 21 invited lectures and 32 contributed papers. A special Technical Exposition was held in the poster session where representatives from MIT, Grenoble and other high field facilities were invited to give a descriptive review of each laboratory. This enabled participants to have a comparative view of the major high magnetic field facilities in the world.

As reflected in these proceedings a wide variety of high field generation methods and material systems were presented in the Symposium. The main purpose of the Symposium was its magnetism orientation. Of course there have been many high magnetic field meetings in the recent decades; for example, the International Conference on Megagauss Magnetic Field Generation and the International Conference of the Application of High Magnetic Fields in Semiconductor Physics are now well established. Work related to the generation of ultrahigh magnetic fields are collected in the former, while applications to transport phenomena are the main concern of the latter meetings. On the other hand, there has been a growing interest in the specific application of high magnetic field to magnetic materials and a comprehensive meeting devoted to high field magnetism seemed desirable. With this in mind, the Organizing Committee solicited and obtained many novel and highly qualified papers. The papers contained in this book cover the magnetization processes and phase transitions under high fields, magnetic interactions, metals and alloys in high fields, spin and charge fluctuations, magneto-optics and high field magnetic resonances. We believe that most of all of the interesting new problems and results recently obtained in high magnetic fields are to be found herein.

It is to be noted that high field magnetism should not be confined to any special topic area but should include to the most general of physical subjects. For example, the so-called Rydberg-Landau problem given in this book will play an important role in astrophysics because the precise solution of hydrogen-like atoms in high magnetic fields is a prerequisite for the understanding of the magnetic structure of white dwarfs or neutron stars. One might also expect applications of high magnetic fields to the biological systems to become one of the important problems in the future.

The Symposium was financially supported by the Japan Society for the Promotion of Science, the Yamada Science Foundation and was also sponsored by the High Magnetic Field Laboratory, Osaka University. The Commemorative Association for the Japan World Exposition gave support through the International Conference on Magnetism-1982-Kyoto. We thank the scientists who served as our program advisors and those who were kind enough to act as session chairmen during the Symposium. We are particularly grateful to our many diligent authors and excellent reviewers whose participation has insured the high quality of these proceedings. Special thanks are due to Dr. Pieter S.H. Bolman and Mrs. Inez van der Heide of North-Holland Publishing Company for their help with the publication process.

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PART 1
INTRODUCTION
TO HIGH FIELD MAGNETISM

ON THE PHYSICS OF HIGH MAGNETIC FIELDS

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A brief list of current areas of research in high field physics is presented covering most of the presentations at this Symposium. More detailed description is given for three topics for which high magnetic fields are required, and which possess unusual interest. These are: 1) Density of states for vibrational states on fractals "Fractons", 2) Thermodynamic properties of exchange enhanced systems, and 3) p-state pairing in thin film or layered superconductors.

1. INTRODUCTION

This Symposium follows at least two others in the rapid development of high magnetic field physics.^{1,2} In addition, a survey, now rather aging, of opportunities for research in high magnetic fields has been prepared.³ The purpose of the present paper is to briefly classify the character of those papers to be presented at this Symposium, and then to describe in outline form three areas which are of particular interest to the author.

The general areas of research in high magnetic fields to be discussed at this Symposium can very roughly be titled as:

- 1) Collective phenomena (e.g., p-wave superconductivity)
- 2) Magnetic structures (e.g., phase transitions, magnetic saturation)
- 3) Atomic-like states (e.g., exciton structure and dynamics)
- 4) Diamagnetism (e.g., orientational ordering of large molecules)
- 5) Thermodynamic properties (e.g., field dependent susceptibilities)
- 6) Transport properties (e.g., quantum oscillations)
- 7) High energy density of states (e.g., vibrations on a fractal)

No list is complete, but this can serve as a rough outline of topics unique to high magnetic field research.

This paper will explore three of the seven areas listed above. The remaining four will be well covered by others at this Symposium. Only one of these three represents original work by this author. However, the significance of the other two warrants some attention.

Each of the three topics is described below in terms of the physical ideas which have been developed, and the possible experimental probes. Space limitations require that the reader be referred to the original treatments for the complete details.

2. DENSITY OF VIBRATIONAL STATES ON A FRACTAL, "FRACTONS"

Fractals are open, self similar structures, with interesting properties as a function of the length scale.⁴ A specific example would be a percolation arrangement where the number of sites on the infinite cluster ($p > p_c$, where p_c is the percolation threshold concentration) increases not as

$$r^d$$

where r is the distance and d the Euclidean dimensionality, but rather as

$$r^{\bar{d}}$$

where \bar{d} is an effective dimensionality, equal to $d - (\beta/\nu)$ in terms of the usual percolation exponents.⁵ This behavior occurs for short length scales in comparison to the coherence length for percolation, ξ_p . For larger lengths one finds usual Euclidean^p properties. If now one examines diffusion along the infinite cluster, the "dead ends" cause a length dependence for the diffusion constant:

$$D(r) \propto r^{-\bar{\delta}}$$

where again for percolation $\bar{\delta} = (t-\beta)/\nu$, t being the conductivity exponent.

The diffusion problem along a fractal can be solved, leading to the ensemble averaged autocorrelation function⁶

$$\langle P_0(t) \rangle \propto t^{-\bar{d}/(2+\bar{\delta})} \quad (1)$$

where the particle has assumed to have been localized at the origin at time $t = 0$.

One now notes that the form of the diffusion equation (Master Equation) is the same as, for example, the harmonic vibrational problem, with a simple replacement of the first time derivative by the second. This mapping allows us to regard the inverse Laplace transform of Eq. (1) as the lattice vibrational density of states

(with ω^2 replacing the Laplace transform spectral parameter ϵ) for a fractal arrangement of masses and springs. One finds

$$N(\omega) \propto \omega^p, \quad p = [2\bar{d}/(2+\bar{d})] - 1 \quad (2)$$

For Euclidean systems, $p = d-1$, so we are led to define, for mode counting purposes, a reciprocal space of effective dimensionality

$$\bar{d} = 2\bar{d}/(2+\bar{d}) \quad (3)$$

We refer to these states, when quantized, as "fractons." Their properties are most interesting. Before we outline them in more detail, some experimental examples are of interest.

Our attention to this problem was aroused by the work of Stapleton et al.⁷ who measured the spin-lattice relaxation time for low-spin Fe(3+) in three hemoproteins. These large molecules were shown by x-ray measurements (counting the increase of the number of alpha carbons with distance for myoglobin at 250 K) to yield a value for $\bar{d} = 1.67 \pm 0.04$, certainly not integral. Their data for the spin-lattice relaxation time as a function of temperature for myoglobin azide (MbN₃) are copied below:

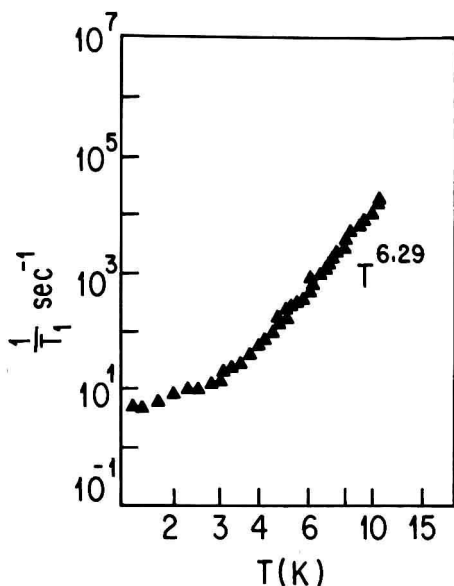


Fig. 1. The electron spin relaxation rate of low-spin Fe(3+) in MbN₃. The rate is fitted to the sum of a direction process, varying as T , and a Raman process, with temperature exponent 6.29.

Their interpretation relied on the use of the usual two-phonon integral for the Raman process relaxation rate, the integrand being proportional to the square of the vibrational density of states. Keeping all other factors the same as for Euclidean space, they extracted the

exponent

$$p = 0.65 \pm 0.04 \quad (4)$$

They did not report other data which would enable us to obtain an independent estimate for \bar{d} . The use of self avoiding random walks as a model for these proteins is inappropriate. For such systems in $d = 3$,⁸

$$\bar{d} = 5/3, \quad \bar{\delta} = 4/3, \quad (5)$$

leading to $\bar{d} = 1$ ($p = 0$), representing one dimensional vibrational behavior.

The essential condition for application of these ideas to physical systems is that the length scale be less than the Euclidean correlation length. For lattice vibrations, this implies that the frequency be greater than a crossover frequency, $\omega_{c.o.}$, itself related to $\bar{\delta}$ by the following expression⁶:

$$\omega_{c.o.}^2 \approx L^{-(2+\bar{\delta})} \quad (6)$$

where L is the size of the fractal object (e.g., the percolation correlation length, or the size of the molecule) in units of the monomer length, and the frequency scale is that of the Debye frequency appropriate to the fractal object. For example, Stapleton et al. state that the temperature range 1 - 20 K is associated with wavelengths of from 10 to 10³ bonds. For a large molecule, this would certainly be consistent with the requirement for fractal behavior.

There are other properties of fractal vibrational states. For example, the vibrational eigenfunctions are local and should not contribute to the thermal conductivity. This behavior (though not with an identification of fractal properties) has recently been reported by Kelham and Rosenberg for epoxy resin, for the energy range of 8 - 50 K (their measurements spanned the range of 0.1 - 80 K).⁹

It is clear that the identification with fractal behavior depends on the condition (6), which then leads to a vibrational density of states (2). The experimental consequences are immediate. The one phonon, or direct relaxation process rate, is directly proportional to the vibrational density of states. If one performs an electron spin lattice relaxation time measurement at sufficiently high magnetic fields, it is possible that one can obtain a direct measurement of the fracton density of states. The field must be sufficiently high that the condition (6) is satisfied. Then the field dependence of the relaxation rate will give the energy dependence of the density of fracton states, and hence a value for p using Eq. (2). The crossover magnetic field will separately give an estimate of $\bar{\delta}$ using Eq. (6). That is, $\bar{\delta}$ is not a free parameter, in that it is determined by the crossover behavior. Finally the factor \bar{d} can also be determined from x-ray measurements, over-determining all the fractal parameters.

There are other interesting consequences of fractal behavior. The eigenstates are supposed to be localized. This could lead to rather interesting magnetic resonance bottleneck effects in that the spatial transfer of excitation will be diffusive rather than wavelike. This might lead to strong bottleneck conditions for both the direct and resonance relaxation processes. Here too, large magnetic fields would be useful to unravel the dynamical properties of a bottleneck. For example, if a bottleneck is found for the resonance relaxation process, the field dependence of the strength of the bottleneck will give a direct estimate of the fracton lifetime (the analysis is similar to that of Gill,¹⁰ but using fractons instead of phonons).

3. THERMODYNAMIC PROPERTIES OF EXCHANGE ENHANCED SYSTEMS

The effect of magnetic fields upon the thermodynamic properties of Fermion systems [e.g., the nonlinear magnetic susceptibility and the field dependent specific heat], depends on the relation between field H and Fermi temperature T_F . Even for extreme fields, this ratio is small (100 Tesla is the equivalent of 170 K in Zeeman splitting). Exchange enhancement caused by electron-electron interactions in a metal can significantly enhance the effect of a magnetic field. A very recent calculation of Béal-Monod and Daniel¹¹ gives a complete analysis of the scaling factors in the presence of spin fluctuations (finite temperature corrections to the $T = 0$ results of Wohlfarth and Rhodes¹²). These can be used to obtain interesting field-induced alterations of the thermodynamics of exchange enhanced systems.

Using a method previously described,¹³ the field scaling factors are found to be:

$$ST/T_F, \quad (7)$$

and

$$S^{3/2}H/T_F \equiv S^{1/2}H/T_{s.f.} \quad (8)$$

Here, S is the Stoner factor,

$$S = (1 - \bar{I})^{-1}$$

where

$$\bar{I} = IN(E_F)$$

We have taken a short-range approximation for the electron-electron interaction, I , and $N(E_F)$ is the density of states at the Fermi energy. In units of the square of the magnetic moment, the Pauli susceptibility (bare) equals

$$\chi_{\text{Pauli}} = 2N(E_F)$$

We have used in Eq. (8) the usual expression for the spin-fluctuation temperature

$$T_{s.f.} = T_F/S$$

Béal-Monod and Daniel give the following expression for the magnetization of an

exchange enhanced Fermi gas (parabolic band) in the temperature and field regimes $T \ll T_{s.f.}$, $H \ll T_{s.f.}/(S)^{1/2}$:

$$M(T, H) = S \chi_{\text{Pauli}} H \left\{ 1 - \alpha_1 S^2 (T/T_F)^2 - \beta_0 S^3 (H/T_F)^2 + (\beta_1 + 4\alpha_1 \beta_0) S^2 (T^2/T_F^2) S^3 (H^2/T_F^2) + \dots \right\} \quad (9)$$

Equation (9) is written in such a way that the scaling relationships are made explicit. For the parabolic band, the coefficients in Eq. (9) equal

$$\beta_0 = 1/6$$

and for $H \ll T$,

$$\alpha_1 = \pi^2/6, \quad \beta_1 = 23 \pi^2/(24)^2$$

while for $T \ll H$,

$$\alpha_1 = \pi^2/4, \quad \beta_1 = 27 \pi^2/(24)^2$$

Some discussion is in order. The $T = 0$ value for β , β_0 , is the value computed in the Stoner-Wohlfarth theory.¹² The temperature dependent contributions to $M(T, H)$ diverge with increasing S . Fluctuations greatly enhance finite temperature corrections to $M(T, H)$. More detailed discussions can be found in the original and complete work of Ref. 11. For more complicated bands (but still isotropic) α_1 and β_0 are known.^{12,14} Béal-Monod and Daniel suggest that α_1 , β_0 , and β_1 will all have the same sign for arbitrary band structure.

Given Eq. (9), Béal-Monod and Daniel¹¹ go on to calculate the field dependence of the specific heat coefficient using a Maxwell relation. For a parabolic band they find

$$\gamma(H) - \gamma(0) = -\chi_{\text{Pauli}} \alpha_1 \left(\frac{S^{3/2}H}{T_F} \right)^2 \times \left[1 - \left(\frac{\beta_1}{2\alpha_1} + 2\beta_0 \right) \left(\frac{S^{3/2}H}{T_F} \right)^2 \dots \right] \quad (10)$$

Béal-Monod points out¹⁵ that the first term in Eq. (10) arises from the curvature of the zero field susceptibility at $T = 0$. At high fields, the second term can contribute significantly, perhaps even reversing the sign of the field dependence of $\gamma(H)$, though of course higher order terms must also be included.

Comparison with experiment is becoming possible with the advent of high field measurements on exchanged enhanced materials. The case of UAl_2 will be analyzed at this Symposium by F. R. de Boer et al., and seems to show the same qualitative behavior as predicted by Eq. (9).

Béal-Monod and Daniel¹¹ have analyzed in some detail the case of liquid He³ where no band structure effects are expected. At the melting pressure, $S \approx 20$, and the departure from linearity of $M(T, H)$ is predicted to be of the order of 2% at 10 Tesla. In confined geometries, however, S can be made as high as 60, giving 25% effects under the same conditions.

The case of Pd continues to be difficult to sort out. The susceptibility at $H = 0$ increases with increasing temperature. This implies, from Eq. (9), that α_1 is negative. This then leads one to expect an increase of the specific heat coefficient $\gamma(H)$ with increasing H using Eq. (10). This seems to be at variance with experiment, though more recent studies do seem to exhibit smaller decreases of $\gamma(H)$ with increasing H than before.¹⁶

The utility of Eqs. (9) and (10) lies with their relationship to one another. The two effects are not independent, and as seen in the example of Pd metal, there is a consistency requirement. One cannot simply introduce arbitrary coefficients for the temperature and field dependences of the susceptibility and specific heat. Rather, the various behaviors are linked through a set of known relationships. This should greatly assist experimental analysis, and may serve as an indication of unwanted impurities present when the consistency relations are not satisfied.

4. P-STATE PAIRING IN THIN FILM OR LAYERED SUPERCONDUCTORS

Use of high magnetic fields to achieve the p-wave condensation state for superconductors has been re-examined recently by Klemm and Scharnberg.¹⁷ There are many problems associated with observation of this state, the most serious perhaps being that ordinary impurity scattering acts as a pair-breaker for p-wave¹⁸ condensation, in strong contrast to the s-wave case where the Anderson theorem shows that the critical temperature is essentially unaffected. Added to this difficulty is the expectation that the p-wave transition temperature, T_T (i.e., triplet), is expected to be much smaller than T_S (i.e., singlet) for s-wave condensation. One argument favoring triplet pairing in a magnetic field is that singlet pairing (clean, type II), is limited by Pauli pair breaking,¹⁹ but triplet pairing is not. Though this be true, Scharnberg and Klemm have recently shown²⁰ that orbital pair breaking in the presence of a field limits triplet condensation in nearly the same manner as for singlet condensation. As a consequence, for bulk materials, unless T_T is very close to T_S (unlikely) orbital pair breaking would prevent the upper critical field H_{c2} for triplet pairing from exceeding the Pauli c_2 limit.

The issue, then, is how to achieve a condition where orbital pair breaking can be suppressed,

in the presence of large enough magnetic field to quench singlet superconductivity.

The idea of using reduced dimensionality for this purpose was first put forward by Efetov and Larkin.²¹ They examined layered compounds [specifically, TaS_2 (pyridine)_{1/2}] where a magnetic field parallel to the $1/2$ layers is able to induce vortices between the metallic layers, and hence "decouple" the layers from one another. This decoupling leads to an enhancement of the upper critical field over the bulk value by (roughly)

$$\sim \ell_{tr} \xi_T(0)/d^2 \quad (11)$$

where ℓ_{tr} is the transport mean free path, $\xi_T(0)$ is the zero-temperature triplet coherence length, and d the thickness of the metallic layer. This enhancement can be sufficient to exceed the Pauli limit, allowing for quenching of the singlet state. Unfortunately, this particular system has been shown to be very dirty, leading Klemm and Scharnberg to question this explanation for the observed very large $H_{c2, ||}$.

In their paper,¹⁷ Klemm and Scharnberg have analyzed the nonlocal Gor'kov gap equations for triplet pairing in the presence of magnetic fields under conditions of reduced dimensionality.

The conditions are stringent (specular surface scattering, clean thin films), but their results can conveniently be summarized by their Fig. 3:

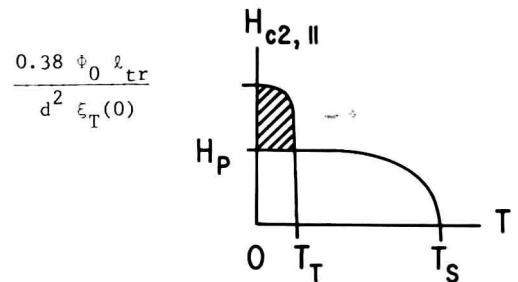


Fig. 2. Schematic plot of $H_{c2, ||}(T)$ for a thin film. The shaded region is $c_2, ||$ the regime of p-wave superconductivity. H_P is the Pauli limiting critical field for s-wave condensation. A similar, though more complicated, curve is exhibited by Ref. 17 for layered compounds.

Klemm and Scharnberg go on to suggest physical systems which might exhibit triplet superconductivity in high magnetic fields. They suggest cleaved thin films of $NbSe_2$, or intercalates of the same system.

One can legitimately ask, beyond the structure of Fig. 2, what signature triplet superconductivity will give which is unique to the p-wave

paired state. Buchholtz and Zwicky have recently pointed out that spin-polarized tunneling (pioneered by Tedrow and Meservey) in a magnetic field appears to be one of the few methods of qualitatively distinguishing its behavior from that of the singlet paired state.²² Another difference would be the effect of magnetic proximity. Whereas a ferromagnetic layer in proximity to a singlet paired superconductor destroys the condensation over a coherence length, quite the opposite is expected to happen for triplet pairing. Indeed, it might be feasible to enhance the effect of an external field by using the proximity effect in conjunction with superposed ferromagnetic layers.

5. CONCLUSION

This paper has not attempted a general survey of the physics that can be done in high magnetic fields. Rather, it has focused on only three areas, in addition to listing those other topics which will be covered in depth by other invited speakers at this Symposium.

The author suggests that the main thrust of the three areas he has discussed are:

- 1) High frequency (infra-red?) electron spin resonance relaxation time measurements, as a function of frequency in the direct process regime (low temperatures, high fields), to probe the "fracton" density of states. Macromolecules, cross-linked polymers, gels, and other "open" self-similar structures would be good candidates for significant departures from the Debye density of states for lattice vibrations.
- 2) High magnetic field studies of magnetization and specific heat of exchange enhanced metals, to determine the thermodynamic parameters α and β . As suggested by Béal-Monod and Daniel, use of He^3 in restricted geometries may be one technique for seeing the nonlinear magnetic field properties of the Fermi gas. Another may simply be use of higher fields and lower temperatures.
- 3) Triplet (p-wave) superconductivity--the existence of the state and its properties. Recent work of Klemm and Scharnberg have given the limits on the magnetic field range (for clean systems, but still type-II) in which the s-wave state is quenched, but the magnetic field is below the upper critical field for p-wave pairing. The use of restricted geometry is essential in order to avoid orbital field-induced pair-breaking for the p-wave state. Thin films and layered compounds are suggested as excellent candidates.

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