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PRESSURE QUENCHED EXCITONIC SOLIDS(I) RENSSELAER
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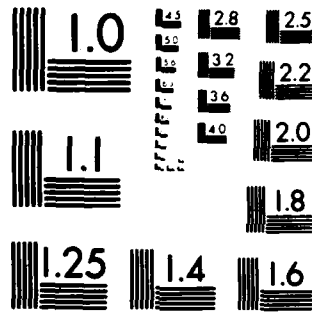
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varying magnetic field so as to reduce the free energy of the system. Such a network is similar to an array of superconducting loops which are orientable although not completely free. If the low field strong diamagnetism is to be attributed to superconductivity, as has been previously suggested we are not yet able to incorporate the high field behavior into such a picture. It is possible that the behavior seen in some samples of cuprous chloride and in cadmium sulfide are due to a not yet understood mechanism.

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ANNUAL REPORT

PRESSURE QUENCHED EXCITONIC SOLIDS

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by

Edmond Brown

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Air Force Office of Scientific Research

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The large diamagnetism associated with pressure quenched cadmium sulfide and copper chloride has suggested the possibility that, at least, some portion of the samples showing anomalous diamagnetism are in a superconducting state.¹ The fact that some samples show this behavior at 77°K, and even at room temperature has suggested that a coupling mechanism between electrons in some solids can bring about superconductivity at anomalously high temperatures. Typically superconductivity is associated with temperatures below 25°K and there is a strong body of thought that suggests superconductivity brought about by electron-phonon coupling is impossible at room temperature¹ in all solids, with the possible exception of solid hydrogen (a phase requiring pressures of several megabars).

The possibility of superconductivity at temperatures much above 25°K is exciting from the theoretical as well as the practical viewpoint. It has been suggested that mechanisms other than just the electron phonon coupling can lead to an indirect attractive interaction between electrons, giving rise to a pairing mechanism that brings about superconductivity. Most of the candidates for such a mechanism have been regarded as unlikely. However excitonic coupling of various types³⁻⁵ have remained an interesting possibility. One of these, a mechanism proposed by Allender, Bray and Bardeen⁶⁻⁷ requires the proximity of two phases in order to get the effect. Essentially conduction electrons in one phase interact indirectly with one another by tunneling into the second phase and virtually exciting an exciton. All direct attempts at producing two such phases in direct contact so as to produce a superconducting interface have failed. Nevertheless the mechanism may be a possible explanation for the diamagnetism in pressure quenched cadmium sulfide in that several phases are simultaneously present in the metastable composite material that results from a pressure quench.

Such a material would possibly contain
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a network of superconducting interfaces and a tendency to keep flux out of certain regions, thereby displaying a diamagnetic behavior such as has been observed on some samples. Nevertheless unless the network of superconductors allows connected paths over a macroscopic domain its conductivity would not directly display any anomalous behavior.

For this reason it is likely that only the magnetic properties of pressure quenched materials would bear strongly on the question of superconductivity. The theorist has a difficult task trying to shed light on the question of whether there is evidence for room temperature superconductivity in the behavior of such samples. In thermodynamic equilibrium only superconductors have diamagnetic susceptibilities anywhere close to that of some of the pressure quenched samples. To the author's knowledge no postulated system consisting of uncorrelated electrons in either a metallic phase or in discrete atomic-like orbitals has been deemed capable of giving rise to such a strong effect. The question of localized orbits is relevant in that the behavior of cadmium sulfide is strongly correlated with its impurity concentration. Thus one cannot rule out orbits localized about such impurity sites.

The problem of characterizing pressure quenched systems is extremely difficult. The difficulties arise from several rather obvious facts: (1) The behavior of pressure quenched materials varies from sample to sample. (2) The behavior of any one sample is time dependent changing its magnetic properties dramatically in the course of several hours. (3) One cannot tell whether some of the time dependent changes are history dependent (affected by exposure to strong fields and varying temperatures.) (4) There is not much knowledge of the arrangement of the various phases which are present in the composite material. Since detailed theoretical work on the microscopic properties of

such a complex system is unlikely to produce meaningful information it was considered better to focus theoretical attention on the broader question of whether the full range of magnetic behavior of pressure quenched cadmium sulfide was compatible with a superconducting model. Up to this point we had been concentrating on the behavior of some of the samples at fields below two kilogauss, where the behavior looked typically like that of certain type II superconductors. On the other hand the behavior at high fields was associated with a strong positive magnetic moment which could not correspond to ordinary paramagnetism. The effect appeared to be somewhat ferromagnetic although because the samples were metastable it was impossible to bring them through complete cycles, or hysteresis loops. In fact the behavior was so history dependent that it suggested the possibility that trapped flux of some sort could be responsible for such unusual behavior. Neither of the principal investigators had encountered any such systems before. The simple idea occurred to us that a superconducting loop containing flux behaves like a magnetic dipole in that it has a magnetic moment associated with it. However, it differs from an elementary dipole in that when it is subject to an external magnetic field it is not the dipole moment of the system that is fixed. Rather the invariant quantity, assuming no flux leakage, is the magnetic flux through the loop. It can be easily shown that such a loop will act like a permanent dipole in a "weak" external field. The word weak implies that the flux provided by the external field is small compared to the total flux in the sample, independent of the orientation. If B_e is the external magnetic induction, and A is the area of a loop, the condition is $B_e A \ll \phi_t$, where ϕ_t is the trapped flux, an invariant quantity. It was somewhat surprising to us that a system of superconducting loops, which were free to turn, would behave like a paramagnetic system in any limit. The question arose as to whether the

magnetic phenomena observed at high fields could in any way be the result of superconducting subsystems. It was not assumed that the superconducting loops surrounding trapped flux required atomic movements on a large scale in order to obtain a turning degree of freedom. Rather, it was postulated that alternative superconducting paths were available for the current so as to simulate the effect of turning. In a primitive model such as this we assume that the effective area of the loop is not sensitive to its orientation. A model such as this is capable of explaining some of the history dependent phenomena in the low magnetic field region but it failed to explain any of the ferromagnetic behavior observed around ten kilogauss.

The strange behavior of cadmium sulfide would not be entirely explained in terms of a superconducting model since that would leave the high field behavior still unaccounted for. It seems to suggest an entirely new mechanism unless one postulates one mechanism in one regime (superconductivity at low magnetic fields) and another mechanism in another (ferromagnetism at higher fields). Such an assumption raises more questions than it settles.

During the academic year covered by this report a research assistant was hired to examine some phenomenological aspects of superconductivity, as described by the London and the GLAG theories (Ginsburg, Landau, Abrikosov and Gorkov), to see whether any of the magnetic phenomena already mentioned could be accounted for. The results were all negative.

This report does not contain any reference to the experimental work carried out under the direction of Dr. R. MacCrone during the year covered. This represented a considerable portion of the research supported by this contract. It is expected that Dr. MacCrone will report on this separately.

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