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Superconductivity in Lead-Base Solid Solution Alloys*

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Magnetization as a function of applied magnetic field has been measured at 4.2°K for binary solid solution alloys of In, Tl, Sn, Hg, Na, and Bi in Pb. For pure Pb, superconductivity is destroyed when the field penetrates abruptly at the thermodynamic critical field H_C. With sufficient alloying element in solution, the field, instead, first penetrates at $H_{FP}(<H_C)$ and penetrates gradually, full penetration and destruction of superconductivity not being reached until H_N $(>H_C)$. Increasing solute concentration decreases H_{FP} and increases H_N in a manner predicted by the "negative surface energy" theories of Abrikosov and others. Annealed specimens approach reversible magnetic behavior and exhibit little trapped flux. Plastic deformation is found to increase H_{FP}, the area under the ascending magnetization curves, the hysteresis, and the trapped flux, but has little effect on H_N. The results indicate that the high critical field of these alloys results from the lowered surface energy of a superconducting-normal interface and not from any specific filamentary features of the microstructure. Dislocations do, however, increase the magnetic hysteresis and, it is inferred, the current-carrying capacity of these alloys.

* For complete article, see J. D. Livingston, Phys. Rev. **129**, 1943 (1963).

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Heusler Alloys*

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The results are given of comprehensive measurements on the magnetic and crystallographic properties of the ferromagnetic Heusler alloys Cu_2MnAl, Cu_2MnIn, Cu_2MnSn, Cu_2MnGe, Au_2MnAl, and the antiferromagnetic alloy Cu_2MnSb. The alloys Cu_2MnBi and Cu_2MnGa are polyphase and the ferromagnetism may be due to binary phases present. The indirect exchange effects which may give rise to ferromagnetism and antiferromagnetism in Heusler alloys are discussed.

INTRODUCTION

THE Heusler alloys have been of interest ever since the discovery over sixty years ago[1] of the first of the series Cu_2MnSn because, although the component elements are paramagnetic or diamagnetic, the alloy itself is ferromagnetic. A comprehensive crystallographic investigation of the structure of this alloy has been carried out by Bradley and Rodgers[2] on Cu_2MnAl. Other Heusler alloys with Al replaced by elements in group III, IV, or V have since been reported. The structure of the ordered beta-phase alloy is bcc with copper at the cube corners and Mn and Al at alternate body centers; the lattice parameter of the alloys $2a_0$ is about 6 Å and the Mn–Mn nearest neighbor distance $(2a_0\sim 4.2$ Å). The maximum saturation magnetization corresponds to a moment of about 4 Bohr magnetons per manganese atom, assuming it is all concentrated on that atom. The reason for the ferromagnetism has been the subject of some speculation but a consideration of the problem has not been assisted by incomplete and inconsistent data. Some of this arises from the fact that the alloy must be quenched from 500–800°C down to room temperature to preserve the high-temperature ordered beta phase and subsequent annealing at an intermediate temperature is necessary; variations in the treatment introduce considerable variations in the ordering. If segregation occurs it may well take place in the form of one of the binary alloys of manganese with aluminium or its substitute, many of which are themselves ferromagnetic.

EXPERIMENTAL

The alloys were prepared by melting in argon at a few millimeters pressure, to prevent evaporation, and homogenised in the temperature range 500–800°C before quenching to room temperature. The quenching temperature (Q.T.) given in Table I is that giving the maximum saturation magnetization. Annealing was carried out at about 200°C for several days to produce an alloy of optimum order; this was checked from Debye–Scherrer analysis and from the effect on the saturation magnetization which increases with increasing order. The saturation magnetization was measured using the standard Sucksmith techniques[3]

* Work supported by European Research Office of the U. S. Department of the Army.
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TABLE I. Table of properties of the Heusler alloys. 2 a_0, lattice spacing (Å). Mn–Mn, nearest neighbor distance of Mn atoms (A); $\sigma_{0,0}$, specific magnetization extrapolated to 0°K emu/gm^{-1}; M, magnetic moment in Bohr magnetons per Mn atom; θ_f, Curie temperature; Q.T., quenching temperature.

	2 a_0	Mn–Mn	$\sigma_{0,0}$	$M\,(\mu_B)$	θ_f	Q.T.
Cu_2Mn Al	5.949	4.21	110	4.12	630°K	650°C
Cu_2Mn Sn	6.173	4.37	76.4	4.11	?	625°C
Cu_2Mn In	6.206	4.39	74.2	3.95	500°K	535°C
Cu_2Mn Sb	6.096	4.31	...	...	$\theta_N=38$°K	650°C
Cu_2Mn Ge	bc tetragonal $c/a=0.96$	4.9	61	2.84	300°K	800°C
Au_2Mn Al	6.36	4.50	20.6	2.24	200°K	700°C
Cu_2Mn Bi, Cu_2Mn Ga	Polyphase					

and extrapolation of the (σ, T) curves was carried out using at $T^{\frac{3}{2}}$ or T^2 law. In most cases several samples of an alloy were prepared with composition varying slightly from the formula, Cu_2MnX, but the differences in σ were not significant.

RESULTS

Cu_2Mn Al[4]

A slight increase in order was produced by annealing at 200°C for some days; the value of σ is thus higher than the previously reported value.

Cu_2Mn Sn[5,6]

The Curie temperature could not be measured because of irreversible precipitation effects which occurred as the temperature was increased; the precipitation was sufficient to be observed by ordinary metallographic examination as well as by x-ray powder methods. The reduction in σ with temperature is associated with this phase change rather than with a true (σ, T) effect and it is probable that the value of θ_f, given in earlier work is associated with this phase change.

Cu_2Mn In

The results are in close agreement with those of Coles, Hume-Rothery, and Myers.[7]

Cu_2Mn Sb

This alloy, which has previously been reported as being ferromagnetic,[8] is found to be paramagnetic at room temperature. The susceptibility increases with decreasing temperature to a maximum at 38°K, suggesting that the alloy is antiferromagnetic below this temperature.[9] A specimen with slightly different composition has the same characteristics. Presumably earlier results had been caused by the presence of the ferromagnetic MnSb.

Cu_2Mn Ge

The structure of these alloys appear to be tetragonal rather than cubic but analysis and indexing of the powder photographs has been difficult. There do not appear to be any earlier reports of this alloy and it is not certain that even with prolonged heat treatment that the bcc phase will be formed.

Au_2Mn Al

Morris, Preston, and Williams[10] reported that this was a ferromagnetic alloy, although they did not measure σ; their value of the lattice constant agrees with the value given in Table I. The results for alloys of slightly different composition are in agreement; the maximum increase brought about by annealing at 200°C was about 5% and it would require some substantial change in the method of preparation to bring the value up to $4\mu_B$. X-ray analysis shows the long range order of the Mn and Al atoms to be incomplete.

Cu_2Mn Bi and Cu_2Mn Ga[11]

These alloys have previously been reported as ferromagnetic. Attempts to produce ordered Heusler alloys have failed and all the alloys prepared were polyphase; the ferromagnetism ($\sim 2\mu_B$ per Mn atom) could be due to the precipitation of binary alloys.

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DISCUSSION

There appears to be general agreement that in the copper alloys the magnetic moment of $4\mu_B$ is concentrated on the Mn atom and this has been confirmed by neutron diffraction experiments on Cu_2MnAl [Wollan and Felcher (private communication)].

The problem of the interaction responsible for the alignment of the spin moments has in the past usually been discussed in terms of the Slater suggestion that ferromagnetism occurs only for a range of value of the ratio R/r of atomic radius R to the effective radius of the d shell r, (Coles, Hume, Rothery, and Myers[6]). However with the large Mn–Mn spacing the direct exchange interaction must be small, and any theory must also account for the paramagnetism and antiferromagnetism of Cu_2MnSb in which the Mn–Mn spacing is essentially the same as in the ferromagnetic Heusler alloy.

A possible mechanism is that suggested by Zener[12] and developed by a number of workers, that the interaction takes place by means of the polarization of the conduction electrons. Yosida[13] has considered the problem of the interaction between the d spins and the conduction electrons in CuMn alloys and has shown that the spin polarization is concentrated near the Mn ions. The polarization oscillates and rapidly decreases with distance R in the form $(x \cos x - \sin x)/x^4$, where $x = 2kR$ and k is the maximum wave vector; for a system with n electrons in a volume v, k is given by $k^3 = 3\pi^2 n/v$.

The oscillation represents a variation from parallel to antiparallel alignment of the spins and the oscillatory curve has nodes at $x = 4.5$, 7.7, 11.0, 14.1. For an alloy with 1.5 conduction electrons per atom (Cu_2MnAl) the nearest neighbor distance corresponds to $x = 7.9$ and for 2 electrons per atom (Cu_2MnSb) $x = 8.7$; both of these values represent ferromagnetic coupling. The next nearest neighbor positions give $x = 11.2$ and 12.2, respectively; the first of these is close to a node and is only slightly antiferromagnetic (Cu_2MnAl) whilst the second (Cu_2MnSb) is strongly antiferromagnetic.

It is not suggested that this is in any way an exact quantitative evaluation of the interaction but it does give an indication of an approach which it is hoped will assist in obtaining an understanding of the problem.

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Superconductors as Permanent Magnets

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A unique property of a superconductor is the possibility of lossless current flow at temperatures below the critical temperature of the superconductor. Hence it is possible to make permanent magnets since fields will exist because of these persistent currents. The potentialities of superconductors in this respect may be distinguished by considering the supercurrent paths in different topological configurations. Experiments on solid and hollow cylinders of lead are reported and compared to theoretical predictions. While the singly connected soft superconductor is uninteresting as a permanent magnet owing to the Meissner effect of flux exclusion, the multiply connected sample has at least some theoretical interest in this application. Considerable improvement may be expected when using the hard or high-field superconductor, since several theories of its behavior imply a connectivity of extremely high multiplicity. It is shown that an energy product of over twenty-five million gauss-oersteds is obtained from Nb_3Sn at 4.2°K. One possible approach to understanding this behavior derives from the Mendelssohn filamentary mesh model. A macroscopic theory of this model postulating that a critical current density as a function of field is the basic property of high-field superconductors is shown to be capable of accounting for the observations. It can be predicted on this basis that other materials or other experimental conditions will make possible much higher values of energy product than reported here.