whereas, from measurements of the width of the magnetic phase mismatch, it is found that a fractional change of $\sim 10^{-5}$ is required to suppress the nonlinear interaction by a factor $\frac{1}{2}$. This represents a phase change (at ω_3) of ~2 radians over the total interaction distance.

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SUPERCONDUCTIVITY BELOW 20 MILLIDEGREES

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A $Nb_{0.30}Mo_{0.70}$ alloy sample has been found to be a superconductor with a zero-field transition temperature of 0.016°K. At the lowest temperature attained, the stray magnetic field of the laboratory (1.5 gauss) was sufficient to suppress superconductivity. Only when a μ-metal case was used to shield the sample from this stray magnetic field was superconductivity observed. The transition was detected by means of a dc mutual inductance method in which a measuring field of 0.108 gauss was used.[1]

This particular alloy was investigated because earlier data[2] down to 0.05°K indicated that superconductivity either vanishes or passes through a minimum within the composition range of 51 to 80% Mo. Specific-heat data[3,4] have shown that the electronic specific-heat coefficient γ for this alloy system passes through a minimum near $Nb_{0.30}Mo_{0.70}$. Thus on the basis of the BCS theory of superconductivity,[5] T_c might be expected to pass through a minimum at this same composition.

The sample was a section, 3 cm long, cut from a cylindrical polycrystalline bar approximately 0.9 cm in diameter. Temperatures below 1°K were produced by the magnetic cooling method. Thermal contact between the coolant salt and the sample was achieved as follows: Potassium chrome alum was allowed to crystallize about a system of 600 AWG 41 Formvar-coated copper wires to form a cylindrical salt pill 5 cm in length and 1 cm in diameter. The sample was then tightly enmeshed in these wires about 7 cm from the near end of the salt pill with nylon thread and GE adhesive 7031. A carbon resistor was similarly enmeshed in the wires about midway between the salt pill and the sample.

The magnetic susceptibility of the salt pill, as well as that of the sample, was measured by a dc mutual inductance method.[1] Susceptibility measurements on the salt allow one to define a magnetic temperature T^* which, in turn, is converted[6] to the temperature one would measure with a spherical salt pill (T_S^*) by the relation $T^* = T_S^* - \Delta$, where Δ in our case was taken to be 0.020°K. The data of Daniels and Kurti[7] were employed to convert T_S^* to thermodynamic temperature.

After the initial demagnetization, no superconductivity was observed even though the salt cooled to temperatures below that corresponding to a maximum in its susceptibility. (Potassium chrome alum attains its maximum susceptibility at 0.011°K.[7]) Immediately after a subsequent demagnetization the entire Dewar assembly was lifted out of the Bitter solenoid and placed in a μ-metal case. Figure 1 shows the behavior of both the susceptibility of the alloy and the temperature of the salt as functions of time after demagnetization. A superconducting to normal transition is clearly evident at approximately 50 minutes after demagnetization.

The thermodynamic temperature of the salt at the midpoint of this transition is 0.013°K. There is, however, considerable uncertainty involved in deducing the temperature of the sample from the temperature of the salt.[8] In this work, sample temperatures were calculated from the formula[8]

$$\dot{Q} = B[T_h^3 - T_s^3]A, \qquad (1)$$

where $\dot{Q}$ is the heat leak into the copper wire system in erg/sec, A is the copper to salt contact area, T_s is the measured temperature of the salt, T_h is the temperature of the wires (this is to be calculated), and B is an experimentally determined parameter. Data of Daniels and Kurti[7] yield the result that the change in enthalpy of potassium chrome alum from 0.011°K to 0.014°K is 80 erg/g. The salt pill contained 6.86 grams (calculated) and the time required for the temperature of the salt to increase from 0.011°K to 0.014°K was 20 minutes. These data lead to a value for the average total heat leak into the system of 0.46 erg/sec. In the arrangement used the contact area between the salt and the copper wires, A in Eq. (1), was calculated to be 75 cm^2.

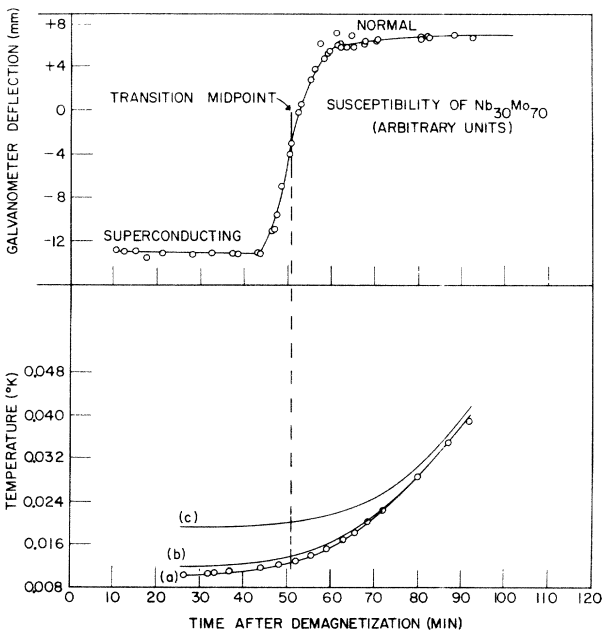

FIG. 1. Variation of several experimental quantities as functions of the time after demagnetization. The uppermost curve shows galvanometer deflections (θ) for the $Nb_{0.30}Mo_{0.70}$ alloy. Changes in θ are directly proportional to changes in the susceptibility χ of the alloy. Curve (*a*) shows the thermodynamic temperature of the salt. Curve (*b*) shows T_w, and curve (*c*) shows T_{sp}. See text for explanation of T_w and T_{sp}.

In order to utilize these data in Eq. (1), we must assign a value to the parameter B. For a slurry of potassium chrome alum, glycerol, and water in contact with insulated copper wires, Spohr[9] found that his data over the temperature interval of 0.013°K to 0.025°K could be adequately represented by Eq. (1) with B equal to 10^3 erg sec^{-1} deg^{-3} cm^{-2}. Wheatley, Griffing, and Estle[10] found at somewhat higher temperatures that single crystals of salt glued to copper plates yielded a B of 2.6×10^4. Using $\dot{Q} = 0.46$ erg/sec and 10^4 and 10^3 for B in Eq. (1), we derived curves *b* and *c* in Fig. 1.

Some part, possibly most, of the total heat leak into the salt pill enters directly through the supports so the use of the total heat leak for $\dot{Q}$ in Eq. (1) leads to an upper limit for the temperature of the sample. Thus it is fairly certain that the temperature of the sample corresponding to the midpoint of the transition shown in Fig. 1 is not greater than 0.020°K. If one takes, as a more realistic value for the $\dot{Q}$ in Eq. (1), half the total heat leak into the system, the two values of B used above lead to a midpoint temperature of either 0.0136°K or 0.0174°K. Temperatures are given to four places; however, the fourth figure has significance only when relative temperatures are discussed. In the remainder of this paper, we always use a $\dot{Q}$ of 0.23 erg/sec and denote temperatures based on a value for B of 10^3 as T_{sp} and those with a B of 10^4 as T_w. Lacking any knowledge of which value of B is more appropriate for the present experimental arrangement, we report the transition temperature to be simply 0.016°K ± 0.003°K.

Figure 1 shows that the normal-state susceptibility was observed when the temperature of the salt was greater than 0.0150°K. Thus the temperature of the sample was either $T_{sp} = 0.0186$°K or $T_w = 0.0154$°K. The width of the transition in Fig. 1, for a measuring field of 0.108 gauss, is

about 0.005°K or one third of T_c itself. A separate experiment conducted with a Sn sample of the same shape and size as the alloy sample yielded the same change in deflection, within experimental limits, when passing from the superconducting to the normal state. Thus the transition in Fig. 1 is consistent with perfect magnetic shielding over the entire volume of the sample.

Perfect magnetic shielding is not a sufficient observation to allow one to conclude that the entire volume of the sample participates in the observed superconducting transition.[11] However, we believe that the transition depicted in Fig. 1 is characteristic of the $Nb_{30}Mo_{70}$ alloy. The particular sample used in the present work is the result of an attempt by H. Sell of the Westinghouse Lamp Division, Bloomfield, New Jersey, to grow a large single crystal of this alloy. While this aim was not realized, microstructure studies of the resulting polycrystalline bar indicated that the bar was comprised of large single-phase grains with no evidence of precipitates along the grain boundaries or in the matrix itself. A quantitative spectroscopic analysis indicated that the sample contained about 5 ppm of Fe.

In an attempt to make some estimate of the magnitude of the critical magnetic fields of this alloy, the susceptibility (χ) was investigated as a function of the magnitude of the measuring or "pipping" field. That is, χ was measured with a "pipping" field corresponding to 1.0 mA through the primary winding, then with 2.0 mA through the primary, then with 3.0 mA, etc. The underlying idea is that as long as χ remains constant the galvanometer deflections will be a linear function of the measuring field. Thus one should obtain one linear dependence for the superconducting state and another for the normal state. Observed galvanometer deflections as a function of the "pipping" field are presented in Fig. 2. This graph is interpreted to indicate that the sample was in the intermediate state for the fields in excess of 0.22 gauss and that the sample was in the normal state for fields in excess of 0.98 gauss. The completion temperature for a critical magnetic field of 0.98 gauss is calculated to be either $T_{sp} = 0.0168$°K or $T_w = 0.0126$°K.

Using the above completion fields and temperatures leads to values for the initial slopes of the critical magnetic field curve for this alloy of -480 gauss/deg (from T_{sp}) and -309 gauss/deg (from T_w). The calorimetric value for γ, 1.46 mJ/mole deg^4, leads to an initial slope of about -195 gauss/deg.

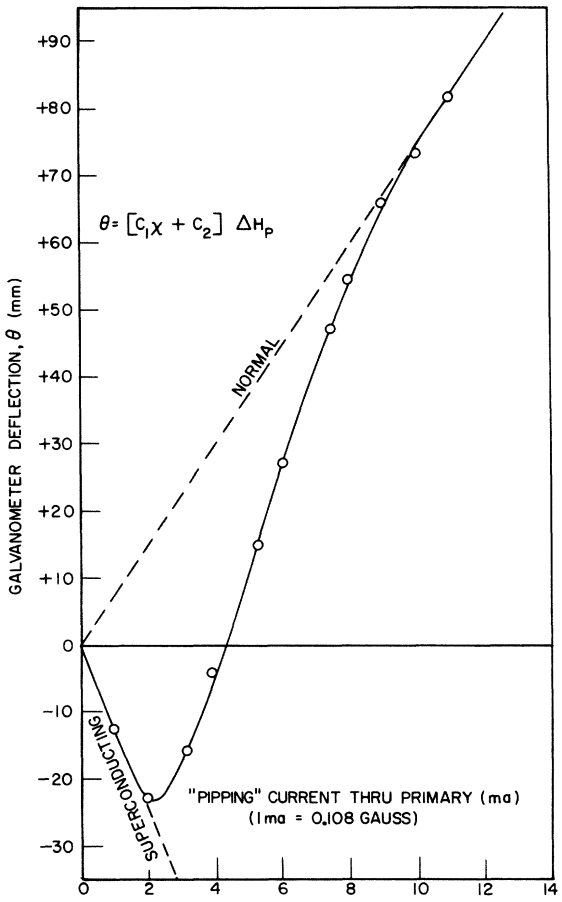

FIG. 2. A plot of the galvanometer deflections of the NbMo alloy as a function of measuring or "pipping" field, ΔH_p. During the time of these measurements the temperature increased by 0.0004°K.

The carbon resistor indicated that the sample attained its lowest temperature, $T_w = 0.012$°K, approximately 25 minutes after the demagnetization. Assuming a parabolic critical magnetic field ($H_0 = 2.7$ gauss and $T_0 = 0.016$°K), our failure to observe superconductivity in a field of 1.5 gauss is an expected and explainable result.

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BIQUADRATIC EXCHANGE BETWEEN Mn^{2+} IONS IN MgO

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Paramagnetic resonance measurements[1] on pairs of Mn^{2+} ions in MgO have indicated values for the isotropic Mn-Mn exchange interactions which are surprisingly large compared with those estimated from bulk measurements on MnO. We have therefore made a more detailed investigation of the pair spectrum. The results suggest that there is an appreciable biquadratic contribution to the exchange of the form $-j(\vec{S}^a\cdot\vec{S}^b)^2$ in addition to the usual bilinear term, $J(\vec{S}^a\cdot\vec{S}^b)$. This not only helps to explain the discrepancy, but may also have significant effects on the magnetic properties of MnO.

The measurements have been made on crystals of MgO containing about 1 at.% Mn using wavelengths 0.85 and 1.25 cm in the temperature range 4 to 200°K. Only nearest-neighbor pairs have been examined in the present experiments, i.e., Mn^{2+} ions occupying lattice sites such as (0, 0, 0) and (1/2, 1/2, 0). Following previous work,[1,2] we first assume that the isotropic exchange is of the usual form, $J(\vec{S}^a\cdot\vec{S}^b)$, where $S^a = S^b = 5/2$ for Mn^{2+} and J is positive (antiferromagnetic). This gives total spin states $S = 0,1,2,3,4,5$, and the interval rule describing the spacing between adjacent states is

$$W_{S,S-1} = JS, \qquad (1)$$

where S is the spin of the higher state.

We have confirmed previous results[1] on the $S = 1$ state and have also identified all the expected transitions belonging to $S = 2$, 3, and 4 from the positions and anisotropies of the lines in the spectrum. (The spectrum from $S = 5$ is obscured by stronger absorption due to lower S states and due to isolated Mn^{2+} ions.) These positions are found to be accurately predicted by anisotropic interaction parameters of the expected form.[2] Having identified the transitions in this way, the energy intervals predicted by Eq. (1) have been investigated by measuring, as a function of temperature, the intensity of transitions belonging to different total spin states both relative to each other and relative to standard comparison substances. The results are given in the first two columns of Table I. It can be seen that the expected interval rule [Eq. (1)] is not obeyed. Much better agreement with experiment would be obtained if the isotropic exchange were of the form

$$\mathcal{H}_{ex} = J(\vec{S}^a\cdot\vec{S}^b)^2 - j(\vec{S}^a\cdot\vec{S}^b)^2, \qquad (2)$$

which gives a modified interval rule

$$W_{S,S-1} = JS - jS[S^2 - S^a(S^a+1) - S^b(S^b+1)]. \qquad (3)$$

Table I. Comparison between predicted and observed Landé intervals.

Measured interval	Experimental energy (°K)	Modified interval rule	Calculated for $j/J = 0.05$ ($J/k = 14.6$°K, $j/k = 0.73$°K)
$\frac{1}{4}\times W_{4,3}$	17.5 ± 3	$J + 1.5j$	15.7
$\frac{1}{3}\times W_{3,2}$	20.0 ± 3	$J + 8.5j$	20.8
$\frac{1}{2}\times W_{2,1}$	24.5 ± 2	$J + 13.5j$	24.5
$1\times W_{1,0}$	28.0 ± 3	$J + 16.5j$	26.7