[15]A review of the relevant experimental data on the electronic state of cerium is given by K. A. Gschneidner, Jr., and R. Smoluchowski, J. Less-Common Metals 5, 374 (1964).

COMMENTS ON "DIRECT EVIDENCE OF FLUX MOTION"

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Pearl[1] has suggested two experiments to demonstrate that potential differences in superconductors are due to moving vortices. One of these experiments employing magnetically coupled films had already been done independently by Giaever[2] and Solomon.[3] Pearl chose the experiment of moving a region of magnetic field in a superconductor in such a way as to produce a net flow of vortices across the sample with no net change in magnetic flux linking the voltmeter circuit. The purpose of this note is to point out that the voltage which Pearl observes is not necessarily due to the motion of vortices and that exactly the same result can be achieved using any magnetoresistive material. Volger and co-workers[4] have pointed out that many effects in superconductors such as their superconducting homopolar dynamo are observable in magnetoresistive materials. There is little doubt that the resistivity of the magnetic field region is due to the motion of vortices when that region is in the mixed state, but unfortunately, Pearl's experiment cannot distinguish this situation from the one in which the region is entirely normal.

If we connect a voltmeter to two points on a sample in which completely general current patterns may be flowing, and if the only source of emf is a time-varying magnetic field, then Kirchhoff's rule may be written

$$\oint \frac{-d\vec{A}}{dt}\cdot d\vec{l} - \oint \frac{\vec{j}_{\text{sample}}}{\sigma}\cdot d\vec{l} = I_m R_m = V, \qquad (1)$$

where $-dA/dt$ describes the time variation of the magnetic field ($\vec{B} = \operatorname{curl}\vec{A}$), $\vec{j}_{\text{sample}}$ is the current density at each point along the path integration (not including the current flowing through the voltmeter), and σ is the conductivity (assumed isotropic) of the sample at each point along the path of integration. It is assumed that the resistance of the meter, R_m, is much greater than the resistance of the sample and leads. I_m is the current through the meter, and V is the voltage.

Consider the two-dimensional analog of Pearl's experiment shown in Fig. 1. For simplicity, we will not consider the boundaries of the sample but it can be shown that these do not affect the nature of the result. The induction will be canceled by simply bringing the voltmeter lead back along the surface of the samples from b to a. The simplest field A which gives the desired magnetic field is shown in Fig. 1. When the region in which the field exists is moved to the right with the velocity v, the magnitude of $-dA/dt$ is given by

$$-dA/dt = Bv\cos\theta. \qquad (2)$$

The charge carriers in the sample in region 2 will respond to this dA/dt field by flowing in the direction of $-dA/dt$, giving rise to a current density $j = -\sigma_2 dA/dt$. There will be a backflow of current outside region 2, so that current continuity is preserved and

$$\int_x^{x+l/\cos\theta} \vec{j}\cdot d\vec{l} = -\left[\int_a^x \vec{j}\cdot d\vec{l} + \int_{x+l/\cos\theta}^b \vec{j}\cdot d\vec{l}\right].$$

The $-dA/dt$ terms cancel out, corresponding to the fact that we have no net flux change in the circuit, and the voltmeter reading is given

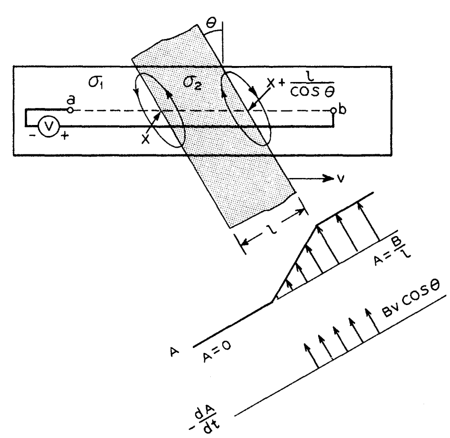

FIG. 1. $\vec{B}$, $\vec{A}$, and $d\vec{A}/dt$ fields in the sample. The induced current patterns are indicated by the loops.

by

$$V=-\frac{1}{\sigma_1}\left[\int_a^x \vec{j}\cdot d\vec{l}+\int_{x+l/\cos\theta}^{b}\vec{j}\cdot d\vec{l}\right]-\frac{1}{\sigma_2}\int_x^{x+l/\cos\theta}\vec{j}\cdot d\vec{l}$$

$$=\left[\frac{1}{\sigma_1}-\frac{1}{\sigma_2}\right]\int_x^{x+l/\cos\theta}\vec{j}\cdot d\vec{l}. \qquad (3)$$

If σ_1 equals σ_2, the voltmeter reading is zero, but if the material is magnetoresistive, and $\sigma_1 \gg \sigma_2$, the voltmeter reading is approximately given by

$$V=+\frac{1}{\sigma_1}\int_x^{x+l/\cos\theta}\sigma_2\frac{d\vec{A}}{dt}\cdot d\vec{l}=+Blv\sin\theta. \qquad (4)$$

Equation (4) indicates that the voltage should be proportional to the velocity, the field, and the width of the region, just as in Pearl's considerations. It should be pointed out that this expression is valid only when points a and b are outside the field region. When θ is increased to the point where these points are included, the voltage will decrease. It would appear, then, that Pearl's experiment indicates only that the conductivity of region 1 is different from region 2 and that the experiment should give the same results in any magnetoresistive material. Moreover, according to this analysis, the voltage in the superconducting should not disappear when the field is increased to the point where region 2 is entirely normal.

Since Pearl's results showed that the voltage disappeared above 6.1°K (the critical temperature for a field of 200 G), it appeared that either the above analysis did not apply to superconductors, or that Pearl's sample was going entirely normal because of heating and/or the fringing of the field. Therefore, we undertook a series of experiments on lead, a lead-indium alloy, and magnetoresistive bismuth to check the validity of the above analysis.

Samples were cut from an approximately 0.005-in.-thick sheet and were approximately 2 mm wide by 3 cm long. They were overlaid by sheets of copper of the same dimensions to cancel out dA/dt. The magnetic field of region 2 was obtained by shaped pole faces on a movable iron-core electromagnet. The angle θ was about 45°. Fields up to 2.5 kG could be obtained. The voltage was amplified and fed into an integrating digital voltmeter. This arrangement could read to 0.01 μV sec, while the observed voltages when the pole irons were moved down over the length of the sample were of the order of 1 μV sec.

When the sample was magnetoresistive bismuth, a voltage was observed. With an alloy of 96% lead and 4% indium ($H_{c2}=1.8$ kG), a small inductive voltage was observed at liquid-nitrogen temperatures because of leads to the sample, but a much larger voltage was observed when the sample was superconducting. This voltage did not disappear at fields greater than H_{c2}. A lead sample exhibited the same behavior but the voltage did not continue to increase when the field was raised well above the critical field of the lead (600 Oe at 4.2°K). In fact, at fields of 1500 Oe, the voltage dropped to about 60% of its maximum value near H_{c2}. It seems likely that flux bundles are being left behind in region 1 and thus decreasing its "conductivity." This would reduce the observed voltage. There is also the problem of heating in the normal region which might change the critical fields, produce thermal emf's, or give rise to any number of experimental complications, which make quantitative experiments difficult. It is significant, however, that a voltage has been observed in bismuth which is approximately the same as one observes in superconductors and that the voltage can be observed in fields up to several times the critical field.

The ideas of reciprocity would lead one to believe that if a current is now passed through the sample, a force should be exerted on the magnet so that it will move in the direction of v. Pearl, in fact, tried to observe this effect. Condon[5] has pointed out that this force would also exist in a magnetoresistive material because the current would be "refracted" at the boundaries of the magnetic field (again we neglect the boundaries of the sample where the current pattern is distorted). If the ratio of the conductivities is high, the current in region 2 must flow perpendicular to the boundary. The magnetic field exerts on the current a force which has a component in the direction of v and if the sample is held fixed while the magnet is free to move in the direction of v, the reaction force should move the magnet.

We have shown that, unfortunately, Pearl's experiment does not provide any more direct evidence of the emf produced by flux motion than the usual resistive-state experiment,[6] since all that one can legitimately conclude is that the conductivity of the region containing the magnetic field is much less than the conductivity of the region where there is no magnetic field.

Discussions with J. H. Condon, Y. B. Kim, and J. Pearl, and the use of the experimental facilities of Y. B. Kim, are gratefully acknowledged.

[1]J. Pearl, Phys. Rev. Letters 16, 99 (1966).

[2]I. Giaever, Phys. Rev. Letters 15, 825 (1965).

[3]P. R. Solomon, Phys. Rev. Letters 16, 50 (1966).

[4]For example, see J. Van Suchtelen, J. Volger, and D. Van Houwelingen, Cryogenics 5, 256 (1965).

[5]J. H. Condon, private communication.

[6]For example, see Y. B. Kim, C. F. Hempstead, and A. R. Strnad, Phys. Rev. Letters 9, 306 (1962).

EFFECT OF ZERO-POINT SPIN DEVIATION ON ENERGY LEVELS OF MAGNETIC IMPURITIES IN ANTIFERROMAGNETS

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In a recent paper Johnson, Dietz, and Guggenheim[1] studied the infrared emission spectra of Ni^{++} as an impurity in the antiferromagnetic crystals MnF_2, $KMnF_3$, and $RbMnF_3$. The ground state of the Ni ion, $^3A_{2g}$, was found to split into three levels, due to the exchange field of the neighbor Mn ions. A small asymmetry in the splitting was observed. We report in this paper more accurate measurements of the asymmetry in the splitting of these levels (see Fig. 1) using unstrained single crystals with smaller concentration of Ni (below 10 parts per million of Mn) and with high resolution. This asymmetry is shown to arise from the effects of the zero-point spin deviation.

For a given crystal, the spacing between the three levels ($M_S = 1, 0, -1$) should be constant according to the molecular-field model, in approximate agreement with the experimental results. However, there are small differences, $\delta E = \Delta E_{1,0} - \Delta E_{0,-1}$, of around 10 cm^{-1} which are not explained within a simple molecular-field model. In order to explain those anomalies, we extend the molecular-field model, by taking into account the difference between the usual effective magnetic field and the actual exchange interaction with neighbors as a perturbation. The Hamiltonian of our system is

$$\mathcal{H} = \beta H \sum_i g_i S_{zi} - 2\sum_{i\neq j} J_{ij}(S_{zi}S_{zj} + S_{xi}S_{xj} + S_{yi}S_{yj}) + \beta H_A\Big(\sum_i g_i S_{zi} - \sum_j g_j S_{zj}\Big),$$

where H_A is the anisotropy field. Usually the exchange interaction J_{ij} is negligible except when i and j are nearest neighbors. The molecular-field theory considers only the first term of the exchange interaction (neglecting the other two) and equates it to an effective magnetic field H_E:

$$\mathcal{H}_0 = \beta H \sum_i g_i S_{zi} + \beta \sum_i g_i H_{Ei} S_{zi} + \beta H_A\Big(\sum_i g_i S_{zi} - \sum_i g_j S_{zj}\Big).$$

Therefore, our perturbation is

$$\mathcal{H}' = -2\sum_{i\neq j} J_{ij}(S_{xi}S_{xj} + S_{yi}S_{yj}) = -\sum_{i\neq j} J_{ij}(S_{+,i}S_{-,j} + S_{-,i}S_{+,j}).$$

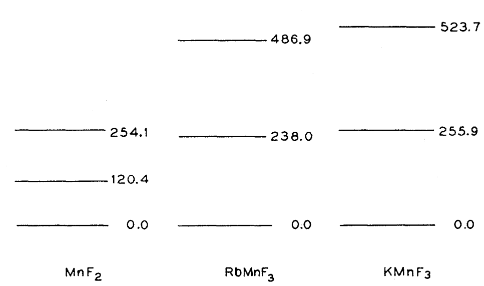

FIG. 1. Energy levels of Ni^{++} in different antiferromagnetic crystals at 4.2°K, in cm^{-1}. The emission lines from $^3T_2$ are about 1 cm^{-1} half-width, and the frequencies quoted above are obtained from splittings measured between the peaks of the lines. The estimated error in measuring the splittings is about 0.1 cm^{-1}.