



FIG. 4. de Haas-van Alphen effect in Ta at 1.13°K. The beating effect due to two oscillating terms can be seen as the field decreases (from right to left) from 141.6 to 134.9 kilogauss during a sweep time of 1.0 millisecond.

tron model^{8,9} the holes in the second band would be expected to give rise to periods shorter than those observed.

Two oscillating terms were also observed in Ta (see Fig. 4), and the corresponding dHvA parameters are listed in Table I. Our measurements indicate that the shorter period term is quite sensitive to orientation, for it nearly disappeared when the sample was removed and remounted at a slightly different angle. It is interesting that Ta and Nb have the same valence and crystal structure, and have lattice parameters differing by less than one percent. For such a situation the

nearly free electron model predicts almost identical periods. The observed differences (less than 50%) could be attributed to the difference in crystallographic orientation.

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FERROMAGNETIC EXCHANGE INTERACTION BETWEEN Mn^{2+} IONS IN Mn, ZnF_2

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Paramagnetic resonance techniques have been used to measure exchange interactions between nearest neighbor (nn) Mn^{2+} ions in mixed crystals of Mn, ZnF_2 with $Mn:Zn \approx 1:50$. The two ions are separated by the short edge ($c \approx 3.2$ Å) of the body-centered tetragonal unit cell. The general procedure has followed that used for previous investigations^{1,2} of exchange-coupled pairs of spins, $S^i = S^j = \frac{5}{2}$.

The results can be described by the following spin Hamiltonians. For one ion of the pair, omitting interactions with the other ion,

$$\mathcal{H}^i = g\beta\vec{H} \cdot \vec{S}^i + D_c [(S_z^i)^2 - \frac{1}{3}S^i(S^i + 1)] + E_c [(S_x^i)^2 - (S_y^i)^2], \quad (1)$$

where $g = 2$, $S^i = \frac{5}{2}$, $D_c = -0.0135 \pm 0.004$ cm⁻¹, E_c

$= 0 \pm 0.006 \text{ cm}^{-1}$. There is a similar Hamiltonian \mathcal{H}^j for S^j . For the interacting pair the Hamiltonian is

$$\mathcal{H}^{ij} = \mathcal{H}^i + \mathcal{H}^j + J \vec{S}^i \cdot \vec{S}^j + D_e [3S_z^i S_z^j - \vec{S}^i \cdot \vec{S}^j], \quad (2)$$

where $J/k = -0.4 \pm 0.2^\circ\text{K}$ and $D_e = -0.051 \pm 0.009 \text{ cm}^{-1}$. The z axis is parallel to c , that is, to the line joining the ions. The isotropic exchange term $J \vec{S}^i \cdot \vec{S}^j$ gives rise to states of total spin ($\vec{S} = \vec{S}^i + \vec{S}^j$) with $S = 0, 1, 2, 3, 4$, and 5 , and with relative energies, $0, J, 3J, 6J, 10J$, and $15J$, respectively. Since J is negative for the present case, $S = 5$ is lowest and $S = 4$ is $5|J|$ higher in energy, and most of the measurements have been made on transitions within these two lowest total spin states.

The values of the parameters D_c and D_e , which determine the position and anisotropy of the lines in the spectrum, have been confirmed by fitting more than 20 transitions in the wavelength range 0.85 to 3.3 cm. The crystal-field contributions, $D_c = -0.0135 \text{ cm}^{-1}$, $E_c = 0$, are of the same order as those found for isolated Mn^{2+} ions in very dilute crystals which are³ $D_c = -0.0186 \text{ cm}^{-1}$, $E_c = -0.0041 \text{ cm}^{-1}$. The other contribution D_e appears to be consistent with the expected dipole-dipole interaction $-g^2\beta^2/r_{ij}^3$, since the value can be fitted by choosing $r_{ij} = 3.21 \text{ \AA}$ which lies between the expected⁴ separations 3.13 \AA for ZnF_2 and 3.31 \AA for MnF_2 .

The magnitude of the isotropic exchange J has been estimated by using intensity measurements to find the separation between different energy levels given by Eq. (2). This has involved the determination of the intensities of a number of lines in the spectrum both relative to each other and relative to a comparison specimen in the temperature range 1.5 to 20°K . All the measurements agree in giving the exchange ferromagnetic in sign and close to the value $J_{\text{nn}}/k = -0.4^\circ\text{K}$. Preliminary measurements have also

been made on next nearest neighbor (nnn) Mn-Mn pairs which give the approximate result $J_{\text{nnn}}/k = 4^\circ\text{K}$ (antiferromagnetic).

This preliminary result for next nearest neighbors is in reasonable agreement with a molecular field interpretation of the bulk properties of the antiferromagnetic MnF_2 as was expected. The ferromagnetic sign of the nearest neighbor exchange, on the other hand, was not expected. If, as seems reasonable, a similar ferromagnetic interaction is present in MnF_2 , then in order to account for the susceptibility properties it may be necessary to assume appreciable antiferromagnetic exchange between further neighbors on the same magnetic sublattice, e.g., third, fourth, sixth, or seventh nearest neighbors.

Finally, it is of interest to note that the nearest and next nearest neighbor pair structure in MnF_2 is in some ways similar to that in MnO but the ratio $J_{\text{nnn}}/J_{\text{nn}}$ is surprisingly different, being about -10 for MnF_2 and $+1$ for MnO .¹ This is not understood, but a possible explanation may be that there is direct ferromagnetic exchange of magnitude a few degrees Kelvin between nearest neighbors in both salts, and that this is the dominant interaction in MnF_2 where the antiferromagnetic superexchange interactions are smaller.

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