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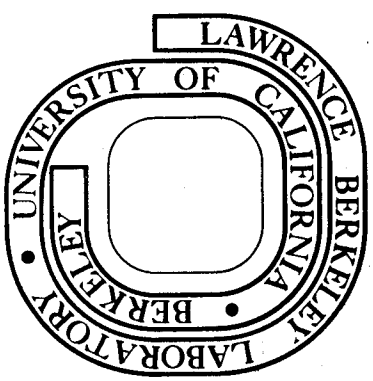

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ABSTRACT

A series of weak absorption lines of unknown origin has been observed below the 8.7 cm^{-1} antiferromagnetic resonance (AFMR) in the far infrared transmission spectra of MnF_2 powders. These extra modes appear in powders from different sources but are not present in the single crystal samples from which powders were prepared. Impurities are therefore unlikely to be the explanation. The strongest mode, at 8.3 cm^{-1}, shows the same magnetic splittings as the AFMR in low fields, yet is apparently not present in 5 cm^{-1} microwave spectra measured at high field values. The integrated strengths of five lines observed between 7.4 cm^{-1} and 8.3 cm^{-1} increase as the particle size is reduced. Surface magnon modes have been predicted in this frequency range. However, the measured dependence on surface area of the 8.3 cm^{-1} mode intensity is not rapid enough to support this interpretation. Microwave transmission spectra show a number of lines below the AFMR field with strengths comparable to that expected for surface modes. No correlation with surface area or with the infrared mode positions has yet been found. Conventional magnetostatic effects cannot account for sharp lines as far from the AFMR as those observed in the infrared spectra.

INTRODUCTION

Manganese flouride has been widely studied as an example of a simple, uniaxial antiferromagnet.[1,2] Its antiferromagnetic resonance (AFMR) occurs at 8.7 cm^{-1} and is accessible to both optical and microwave spectroscopic techniques. In this paper we report the discovery of additional resonances in the far infrared transmission spectrum of MnF_2 powders. A number of possible explanations are considered including the presence of surface spin wave modes. These have been predicted for several crystal structures but not yet observed in any material.

INFRARED MEASUREMENTS

Far infrared transmission spectra were obtained using the techniques of fourier transform spectroscopy. The output of a Michelson interferometer was coupled into a cryostat containing the sample, a high field superconducting solenoid, and a doped germanium bolometer. Powder samples were mounted in cylindrical light pipe sections and saturated in wax to preserve random orientation in magnetic fields. They were immersed in the 4.2 K helium bath.

Figure 1 shows the spectra of five powder samples ground from a single crystal and sieved to segregate grain sizes. The AFMR at 8.7 cm^{-1} appears broad in the 273 mg samples due to magnetostatic modes and to strong saturation of the line. In the 5 mg sample the line is approaching saturation with a 0.15 cm^{-1} linewidth comparable to the instrumental resolution of 0.1 cm^{-1}. Mode strengths for the extra absorptions observed below the AFMR depend strongly on the size of the powder grains. The most intense line at 8.3 cm^{-1} grows stronger as the particle size is reduced. Four other lines located between 7 cm^{-1} and 8.2 cm^{-1} have a similar dependence on grain size but are too weak to be seen except in the finest powder grind. A stronger mode at 7.7 cm^{-1} has a more complicated dependence on particle size. The approximate strengths of these extra modes range from 1/500 to 1/20 of the AFMR strength based upon comparison with the AFMR intensity in unsaturated 1 mg, 5 mg, and 10 mg samples.

The AFMR exhibits a linear magnetic field splitting when the magnetization axis is parallel to the applied field and a smaller quadratic splitting when it is perpendicular. The resultant spectrum for powder samples in an applied field is a broadened absorption with sharp edges on the low and high frequency sides where crystals aligned with the field experience the maximum splitting. For fields up to 14 kOe our spectra show that the splitting pattern of the 8.3 cm^{-1} mode is the same as that of the AFMR indicating its probable antiferromagnetic origin. At higher fields its absorption is too broad to be detected. Studies of the alloy systems $(Co,Mn)F_2$ and $(Fe,Mn)F_2$ also show that the 8.3 cm^{-1} mode is closely related to the AFMR.[3,4] The two lines

merge and the absorption shifts to higher frequencies as the iron or cobalt concentration increases. They are separately visible only for impurity concentrations less than 2 mole percent.

So far no explanation for the extra absorptions has been found which satisfactorily accounts for their position, shape, field dependence, and the variation of absorption strength with particle size. Substitutional impurities in certain antiferromagnetic crystals do cause sharp satellite lines near the AFMR.[3,5] The modes we observe however appear in powder samples from three different sources but are not present in the single crystals from which the powders were prepared. Impurity effects in the original crystals are therefore ruled out. Surface contamination introduced during the grinding process is a possibility but would probably cause broader structure with a different field dependence.

Several other surface and size effects have been eliminated as possible sources of the 8.3 cm^{-1} mode. Interaction of sample crystals with the mounting wax or with each other has been ruled out by preparing a sample without the wax and one with the MnF_2 grains diluted in 10 times the volume of ZnF_2 powder. Both showed the 8.3 cm^{-1} absorption line with the expected intensity. Annealing a powder sample also produced no change in the 8.3 cm^{-1} mode. This makes strains and dislocations introduced during powder grinding an unlikely source of the extra modes. In addition to the 8.7 cm^{-1} AFMR, which corresponds to uniform precession of all spins in a crystal, there exist shape dependent magnetostatic modes[6,7] displaced from the AFMR by as much as 0.13 cm^{-1}. These, along with damping effects due to surface scattering, account for the linewidth of the AFMR but cannot explain a sharp line at 8.3 cm^{-1}.

Finally we consider surface spin waves as a possible source of the modes detected between 7 cm^{-1} and 8.3 cm^{-1}. A free surface in an antiferromagnetic crystal reduces the number of neighboring spins and therefore the exchange forces felt by spins on surface sites. Calculations for different crystal structures and surfaces[8,9] show that the resulting surface magnon frequencies lie in the range 6.2 cm^{-1} to 8.7 cm^{-1} for MnF_2. Predicted penetration depths are typically between 5 and 50 atomic layers and are largest for modes nearest the AFMR. Figure 2 plots the integrated strengths for the 8.3 cm^{-1} line in Fig. 1 against the measured surface to volume ratios (SVR) for four 273 mg powder samples. A surface mode's integrated strength should vary linearly with SVR for very small fractional absorptions. Assuming a

lorentzian line shape this should change to a $\sqrt{SVR}$ dependence for a saturated absorption. The measured variation with surface area is not as rapid as we would expect for a surface magnon mode even in the limit of a fully saturated line. The relative strengths of the surface and bulk modes is the ratio of the crystal volumes participating in the two resonances. This is found by multiplying the SVR of a powder sample by the penetration depth. Figure 1e, where the 8.3 cm^{-1} mode has about 1/50 the AFMR strength and the SVR is 246, requires a penetration depth of at least one micron (2000 lattice constants) for the 8.3 cm^{-1} mode. This is much larger than theory predicts. However the weaker modes appearing below 8.3 cm^{-1} in Fig. 1b do have the intensity expected for surface modes with penetration depth of approximately 10 lattice constants. Optical techniques are not sensitive enough to study the magnetic field or SVR dependence of these lines.

MICROWAVE MEASUREMENTS

Field swept transmission spectra using a 5 cm^{-1} microwave source also show reproducible structure below the AFMR field. However none of it has the expected dependence on surface area and none can be identified with the lines seen in the infrared spectra. For powder samples the microwave experiment detects the linear absorption edge and achieves about the same sensitivity as the infrared data. Surface modes penetrating at least 10 lattice constants are visible in the spectra. Even so the strong 8.3 cm^{-1} mode is not present. This provides evidence that the field dependence observed below 14 kOe does not persist to the 50 kOe field values used to observe the microwave resonance. Microwave measurements on single crystal samples were less sensitive to surface magnons than the powder measurements due to the small surface areas of single crystals.

CONCLUSIONS

None of the possibilities we have considered satisfactorily accounts for the strong 8.3 cm^{-1} absorption detected in MnF_2 powder samples. The weaker lines, visible only in the finest powder, could not be studied as thoroughly. Their intensity, frequency range, and dependence on particle size do, however, suggest surface spin waves as a possible cause. Conclusive proof of this possibility requires more sensitive single crystal spectra in order to identify each mode with a particular crystal surface. Measurements of this type are in progress.

REFERENCES

1. F. M. Johnson and A. H. Nethercott, Phys. Rev. 114, 705 (1959).
2. S. Foner, in Magnetism I, G. T. Rado and H. Suhl, eds., (Academic Press, N.Y., 1963), p. 383.
3. B. Ender, P. L. Richards, and W. E. Tennant, Magnetism and Magnetic Materials 1972 C. D. Graham, Jr. and J. J. Rhyne, eds. (AIP, N.Y., 1972) p. 179.
4. W. E. Tennant, Ph.D. Thesis, University of California, Berkeley, 1974.
5. R. A. Cowley and W. J. L. Burgers, Rev. Mod. Phys. 44, 406 (1972).
6. J. P. Kotthaus and V. Jaccarino, Phys. Rev. Letters 28, 1649 (1972).
7. J. P. Kotthaus and V. Jaccarino, Magnetism and Magnetic Materials 1972 (AIP, N.Y., 1972).
8. D. L. Mills and W. M. Saslow, Phys. Rev. 171, 488 (1968).
9. T. Wolfram and R. E. DeWames, Phys. Rev. 185, 762 (1969).

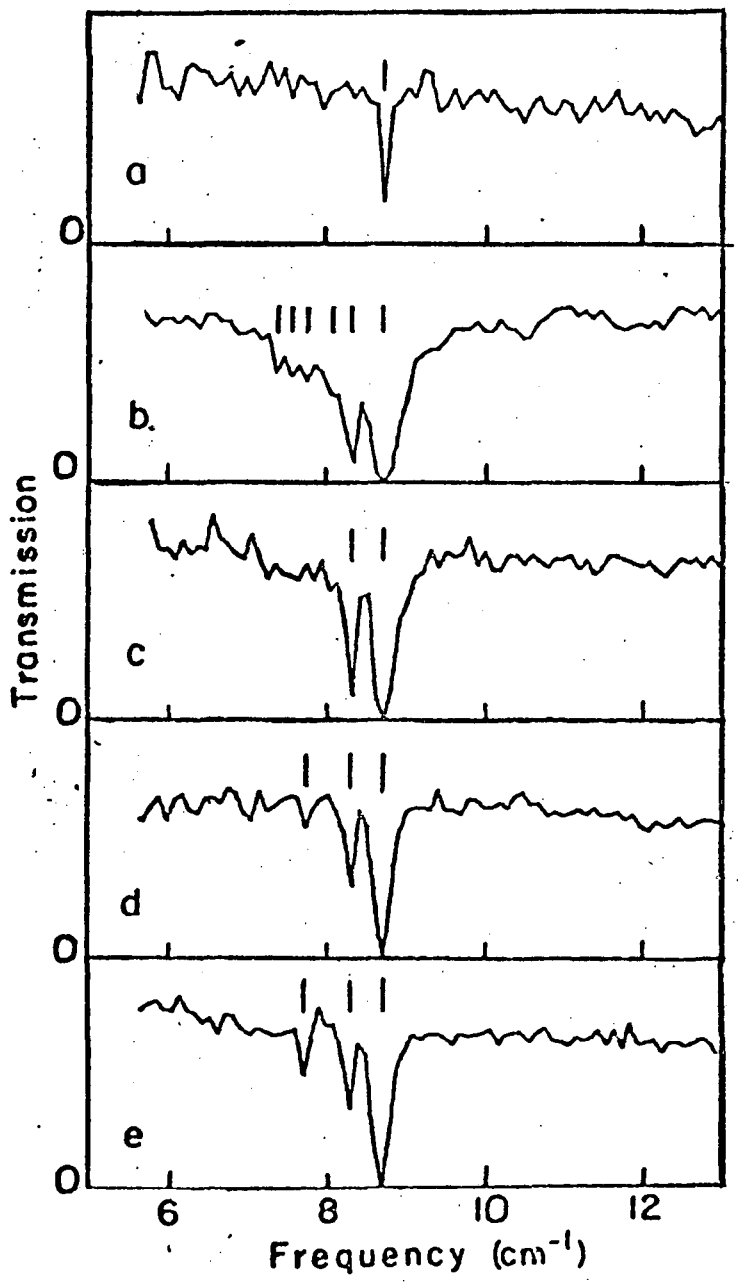

Fig. 1. Transmission spectra of MnF_2 powders. (a) 5 mg sample with surface to volume ratio (SVR) of 50,000 cm^{-1}; (b) 273 mg, SVR=50,000; (c) 273 mg, SVR=1765; (d) 273 mg, SVR=313; (e) 273 mg, SVR=246

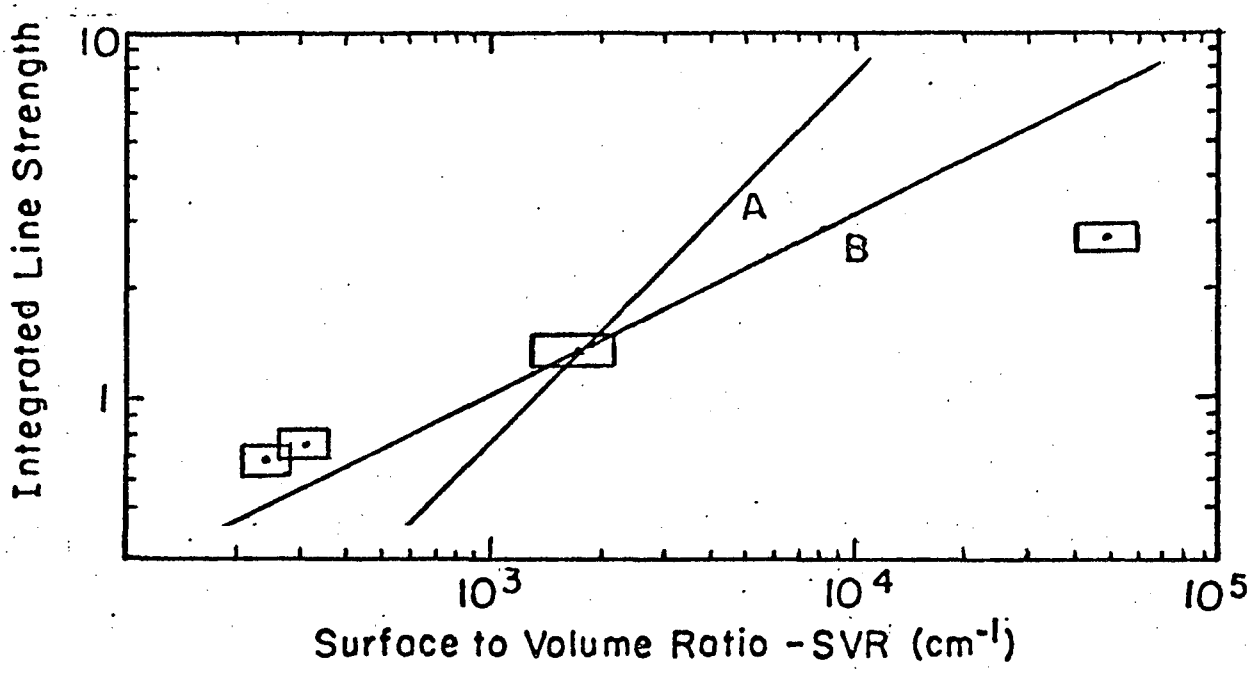

Fig. 2. Logarithmic plot of integrated absorption strength vs. surface-to-volume ratio for the 8.3 cm^{-1} line. Curves A and b show the expected dependence for unsaturated and saturated surface magnon lines respectively.

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