Giant magnetoimpedance near a metal–insulator transition: Study of Fe in a V_2O_3 matrix

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We report microwave absorption measurements as a function of temperature (from 290 to 125 K) and magnetic field (from 0 to 0.3 T) in mm-thick parallelepipeds of sintered V_2O_3 and V_2O_3 containing micron-size Fe precipitates. As before, it turns out that near the metal–insulator (MI) transition, the loss exhibits a sharp peak as a function of temperature. On application of a magnetic field, the peak temperature for (V_2O_3+Fe) changes by a few kelvin, causing a giant magnetoimpedance (≈ 200% in 0.1 T field) in the neighborhood of the MI transition.

It is generally recognized that many useful devices would follow if one could design a material whose microwave loss is sensitive to application of a modest magnetic field. In this connection superconductors,[1,2] amorphous wires,[3] manganite powders,[4] multilayered magnetic films,[5] to name a few, immediately come to mind. In superconductors the field sensitivity is largest in the neighborhood of the critical temperature, presumably because the resistivity drops precipitously. The question arises, could one use the metal–insulator (MI) transition to produce a like effect? This letter describes the results of an attempt at such a material. We started with V_2O_3 (a classic[6,7] MI system), incorporated micron-size Fe particles in it and measured the microwave loss as a function of temperature (T). As we shall see, if one uses mm-size parallelepipeds, the loss exhibits a sharp peak at $T<T_v$ (the Verwey transition temperature). Application of a field moves the peak by a few degrees, thereby producing a giant magnetoimpedance, as large as 200% in a field of 0.1 T. The results can be understood in terms of our previous observations on finite-size samples of $Gd_{0.5}Ba_{0.5}CoO_3$, on the one hand,[8] and micron-size powders of ferromagnetic materials on the other.[9]

$FeVO_4$ was used as the starting material to form the pellets used here. First, $FeVO_4$ was prepared by reacting $FeC_2O_4.2H_2O$ and V_2O_5 in air at 600–850 °C for 30 h. Next, Fe:V_2O_3 samples were obtained by reduction of $FeVO_4$ in hydrogen at 750–850 °C. The process was monitored by noting the weight loss with a thermogravimetric balance (Cahn TG-131 system). Complete reduction to Fe:V_2O_3(FVO) required 18 days at 750 °C (yielding FVO750) and 10 days at 850 °C (giving FVO850). Powder x-ray diffraction patterns (not shown) showed only Fe and V_2O_3 in the final product; the starting material $FeVO_4$ was completely absent. Further confirmation was obtained by transmission electron microscopy (TEM). The results for the FVO850 sample are in Fig. 1 and show that micron-size (∼3-μm-diam) grains of Fe are precipitated out of the V_2O_3. However, one cannot rule out the possibility that a small amount of Fe (∼ few percent) is

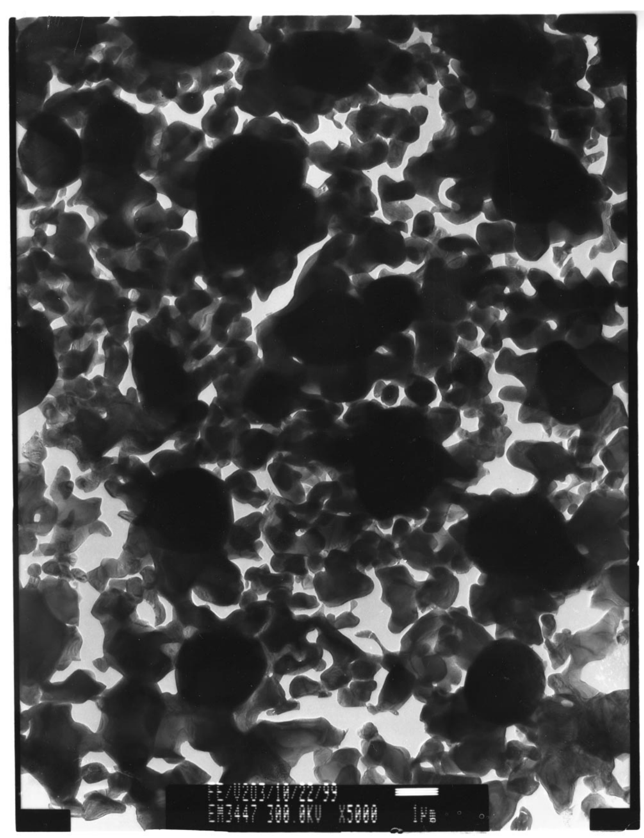

FIG. 1. TEM photograph for FVO850 sample showing 3-μm-diam grains of Fe precipitated out of PVO matrix. White regions correspond to pores in the sample.

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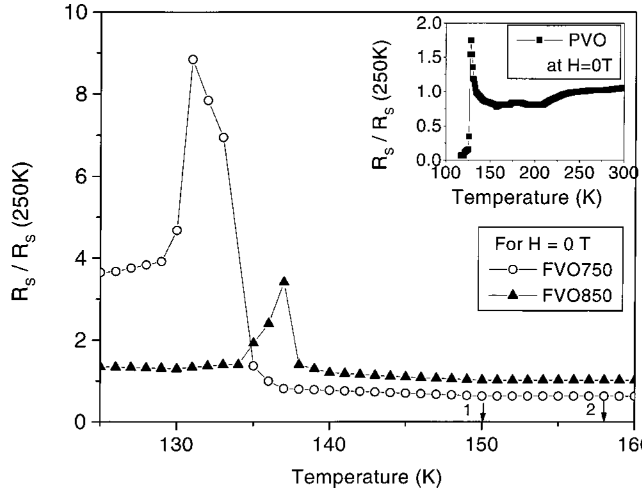

FIG. 2. Zero-field temperature dependence of R_s for FVO samples taken while cooling through the transition region. Arrows 1 and 2 shows the dc metal–insulator transition temperatures in the PVO and FVO samples. The inset shows cooling data for the PVO sample at zero field.

still trapped in V_2O_3. Such a small inclusion is inconsequential, as shown by McWhan *et al.*[7] For the sake of comparison, measurements were also made on pure V_2O_3(PVO) samples. The densities are rather low: 4.1 gm/cm^3 (PVO), 3.5 gm/cm^3 (FVO750), and 5.5 gm/cm^3 (FVO850). Single-crystal PVO has a density of 6.8 gm/cm^3 and Fe, 7.8 gm/cm^3. Thus, the ceramic consists of relatively loosely packed grains. The transmission electron micrographs for FVO750 and FVO850 also show voids (white regions in Fig. 1) with the former having more pores than the latter.

Starting with the disk-shaped (3 cm diam $\times$1.2 mm thick) pellets, samples for different measurements were all cut from a single disk. For microwave studies, they were parallelepipeds, roughly $(1\times0.5\times0.1)$ cm^3. However, the materials tend to crumble, and therefore, the thicknesses are far from precise.

Using the conventional four-probe technique, the dc resistivity was measured as a function of temperature both in zero field and a field of 8.5 T. As in Ref. 9, the microwave losses were studied using the cavity perturbation technique, with the sample being placed in the region of maximum rf magnetic field ($h_{\rm rf}$). For measuring ferromagnetic resonance (FMR), the dc field (H) was applied perpendicular to $h_{\rm rf}$. For magnetoimpedance studies, H was parallel to $h_{\rm rf}$.

At room temperature, the operational[10] resistivities (ρ) of the samples are of the order of 10 mΩ cm. On lowering T, each exhibits a sharp (ρ jumps by three orders of magnitude in $\sim$5 K) MI transition—the transition temperature being about 150 K for PVO (see the arrows in Fig. 2). In single-crystal PVO, $\rho\sim0.3\,\mathrm{m}\Omega$ cm and jumps by six orders of magnitude at T_v.[9]

Addition of Fe moves T_v upwards by about 8 K. These are first-order transitions, exhibiting thermal hysteresis of about 10 K. As expected, application of an 8.5 T field has no effect in PVO. In FVO, there is a very slight, perhaps a few percent, change in resistivity in the transition region. For all practical purposes, the magnetoresistance is negligibly small.

Room-temperature FMR spectra were recorded at 34 and 9.8 GHz. In both FVO samples, the lines are extremely wide (upwards of 0.5 T). Most likely, these large widths are a consequence of the wide variety of grain shapes and orientations. The distribution of shape anisotropy fields (demagnetization effects) has a width of $\sim$1 T, thus the wide lines are not surprising. At the higher frequency, the line center is not too far from $g=2$ (metallic Fe has $g\sim2.09$). At 9.8 GHz, the large linewidth causes a sizable contribution from the nonresonant counter-rotating circular polarization of the rf field, giving rise to a sizable zero-field absorption and shifting the peak absorption down field from $g=2$.

As in Ref. 9, the zero-field loss has two contributions: P_ρ, due to resistive effects, and P_μ, due to the dynamic permeability. Presumably, the latter is responsible for the magnetoimpedance effects discussed below because, as we have seen above, the resistivity is essentially unaltered on application of a magnetic field.

The results of microwave absorption studies are represented here in terms of the surface resistance R_s (normalized to its value at 250 K), which is given by

$$R_s^{\rm expt}=\frac{(\sqrt{r_L}-\sqrt{r_U})(2R_cG)}{(1-\sqrt{r_L}(1+\sqrt{r_U})}, \tag{1}$$

where r is the measure of the power reflected by the cavity (U and L stand for unloaded and loaded cavity), R_c represents wall loss and G is a geometrical factor. Since G is not known with sufficient accuracy, $R_s^{\rm expt}$ is in arbitrary units.[8]

At room temperature, both FVO750 and FVO850 exhibit a reduction in R_s on application of $H\|h_{\rm rf}$, in accord with[9]

$$\frac{R_s(H)}{R_s(0)}=\frac{H^2}{H^2+H_0^2}, \tag{2}$$

where H_0 is the half-point field, $[R_s(H)/R_s(0)=0.5]$. That is, one observes a magnetoimpedance due to the ferromagnetic powder; the Fe inclusions in the PVO matrix. However, the effect is small, $\mu_0H_0\approx0.5\,\mathrm{T}$.

The zero-field temperature dependence of R_s in 1.2-mm-thick parallelepipeds is summarized in Fig. 2. Data were taken while cooling. Sharp peaks are evident. They are located well below the respective dc MI transition temperatures (arrows in Fig. 2). In PVO the effect of thickness was further checked by thinning the sample to 0.5 mm, when the peak moved to 138 K. Qualitatively, these observations are in accord with the findings of Ref. 8. However, as a further check that the MI transition temperature at high frequencies is no different from that obtained by the dc resistivity measurements, a PVO pellet was crushed to yield μm diam powder particles, and the T dependence of the loss was measured. Indeed, the microwave transition temperature agrees with the dc data.

The most telling results of the present investigation are in Figs. 3 and 4, where we show the effect of applying $\sim$0.1 T fields on the peaks observed in the FVO parallelepipeds. It turns out that in a field, the peak in FVO850 (FVO750) moves down (up) in temperature by a few degrees. The magnetoimpedance derived from such data exhibits a huge variation in the transition region, *the largest value being about 200% in FVO750 at 136 K.* It is a clear demonstration of the combined effects of previous findings, namely, (i) a finite-thickness sample exhibits a peak in microwave absorption below the MI transition as the resistivity rises rapidly; (ii) incorporating a ferromagnetic powder makes the peak tem-

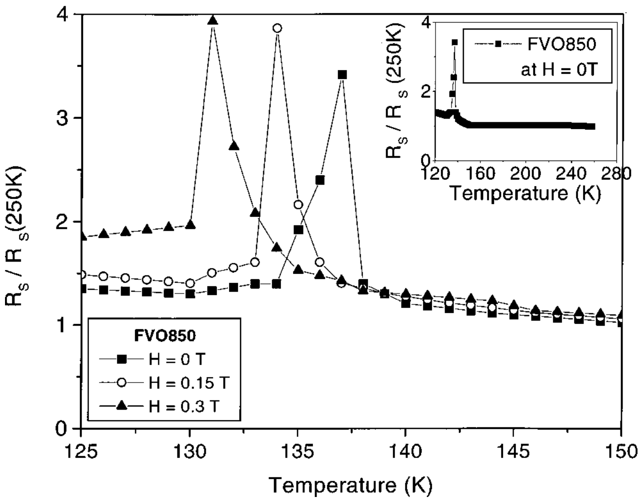

FIG. 3. Effect of magnetic field on R_s peaks for the FVO 850 sample while cooling. It shows the peak moves down from 137 K($H=0$) to 131 K($H=0.3$ T). The inset shows cooling data for the FVO850 sample at zero field.

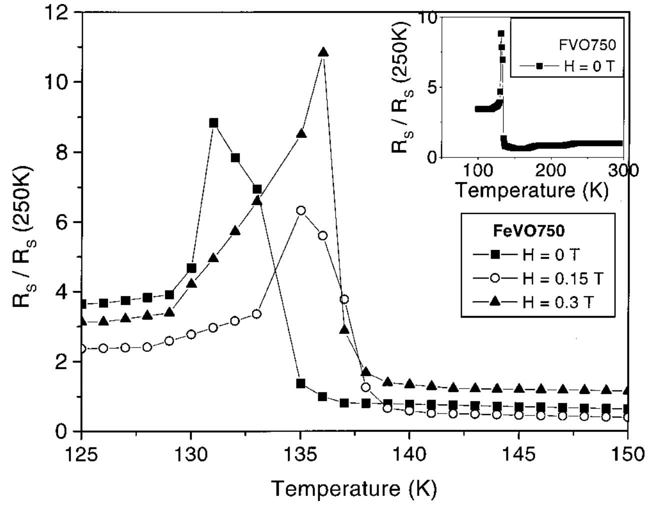

FIG. 4. Effect of magnetic field on R_s peaks for the FVO 750 sample while cooling. It shows the peak moves up from 131 K($H=0$) to 136 K($H=0.3$ T). The inset shows the data at zero field over the full temperature range.

perature sensitive to magnetic field; and (iii) thereby affects the occurrence of a giant magnetoimpedance near the MI transition temperature.

In principle, one can generalize the R_s expression of Ref. 8 to include the effects of absorption in the spin system, by writing for a sample of thickness t,

$$R_s=\left(\frac{\rho}{\delta_0}\right)\frac{\mu^+\tanh(a\mu^+)[1+\tan^2(a\mu^-)]-\mu^-\tan(a\mu^-)[1-\tanh^2(a\mu^+)]}{1+\tan^2(a\mu^-)\tanh^2(a\mu^+)}, \tag{3}$$

where

$$a=(t/2\delta_0), \delta_0=\sqrt{\frac{2\rho}{\mu_0\omega}},$$

$$\mu^+=\sqrt{\sqrt{\mu_1^2+\mu_2^2}+\mu_2},$$

and

$$\mu^-=\sqrt{\sqrt{\mu_1^2+\mu_2^2}-\mu_2}$$

with the dynamic permeability $\mu=\mu_1+i\mu_2$, being a complicated function of both H and T. However, with several adjustable parameters, it does not appear fruitful to attempt quantitative fits, especially when one realizes that the ceramic samples are far from compact and the thicknesses less than precise.

A rough qualitative understanding follows if we consider Eq. (3) in the limit $\mu_2 \gg \mu_1$. Then $\mu^- \sim 0$, $\mu^+ \rightarrow \sqrt{2\mu_2}$ and Eq. (3) reduces to

$$R_s=\left(\frac{\rho}{\delta_\mu}\right)\tanh(a_\mu), \tag{4}$$

with $\delta_\mu=\sqrt{\rho/\mu_0\mu_2\omega}$, $a_\mu=(t/2\delta_\mu)$. This will exhibit a peak for $a_\mu \approx 1$. Thus, if on application of a field μ_2 reduces (increases), the peak will move up (down) in temperature. As noted above, the FMR lines are very wide. At 9.8 GHz, the maximum absorption in FVO750 occurs close to zero field while in FVO850 it is around 0.14 T. Hence, the opposite behavior of the R_s peaks is seen in the two cases.

In conclusion, by combining a material (V_2O_3) showing a classic MI transition and a ferromagnetic powder (Fe), it has been demonstrated that one can design a sample to exhibit a giant magnetoimpedance at microwave frequencies within a specific temperature interval close to the metal–insulator transition temperature. In order to make this discovery more amenable to possible applications, attempts are underway to obtain this effect near room temperature by choosing appropriate materials.

The Department of Science and Technology, Government of India supported the work at Bangalore.

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[10] To obtain the ''resistivity,'' we used the overall sample size. In a sample with large numbers of voids this can, at best, yield an operational value.