Susceptibility of Fe in Cu

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The macroscopic and microscopic susceptibility measurements of Fe in Cu are reexamined. The data as they stand indicate a small antiferromagnetic polarization in the Kondo state.

The understanding of the properties of a local magnetic moment in a metallic environment poses considerable problems.[1,2] If the coupling between the local magnetic moment and the conduction electrons is negative, the Kondo effect occurs; this means a gradual disappearance of the moment below a certain temperature T_K. For the understanding of the nature of the Kondo state it is interesting to see whether this vanishing of the moment occurs by a change of the moment itself or by the buildup of a cloud of antiferromagnetically-polarized conduction electrons around the moment. In this latter case the conduction electrons would contribute an extra contribution to the magnetic susceptibility of the system in the Kondo state. This extra contribution can be extracted by finding a difference between the local susceptibility measured, e.g., by an experiment which uses the nucleus as a probe or a neutron scattering experiment, and comparing these results with the data from a macroscopic magnetization experiment. If these data show a difference, this would strongly suggest a polarization in the electron gas.

Steiner *et al.*[3] recently analyzed Mössbauer-effect data (local susceptibility) and macroscopic-susceptibility data of a typical Kondo system *Cu* Fe. This analysis indicated an extra contribution to the total susceptibility at temperatures below 30 °K.

Campbell[4] challenges this interpretation in the preceding paper. He suggests that the data as they stand can also be explained by a straight line (full line in Ref. 4, Fig. 1) if one only allows a nonzero intercept of the curve indicating an orbital (which is temperature independent and a constant) term to the susceptibility.[5] The problem with the interpretation as suggested by the straight line in Fig. 1 of Ref. 4 lies in the fact that it can only be correct if the errors of the data of Ref. 3 are increased by more than a factor of 5. Taking into account all possible sources of error this seems an unlikely possibility.

In view of the importance of the problem, the available data (macroscopic and microscopic) were reanalyzed. This was done in every case with a computer program, to eliminate a possible bias. The first question is, as is obvious from the work, e.g., of Narath,[5] and also the preceding paper,[4] in how far there is any orbital-temperature-independent contribution to the susceptibility. To that object all the available macroscopic[6-9] susceptibility data were taken and extrapolated as well as possible to zero Fe concentration. This is an important correction because at 10 °K and 100 ppm the pair contribution to the total susceptibility amounts already to 10%. Therefore some of the low-temperature data of Refs. 6 and 9 were omitted because the extrapolation to zero concentration could not be performed with sufficient accuracy.

A curve of the form

$$\chi_{tot} = \alpha/(T+\Theta)+\beta$$

was fitted to the data. This procedure yielded a good fit to all the data,[6-9] with

$$\alpha = 27.9(7)\times10^{-5}\ \mu_B\,\mathrm{K/G}\ ,$$

$$\beta = 1.2(1)\times10^{-7}\ \mu_B/\mathrm{G}\ ,$$

$$\Theta = 27.6(6)\ \mathrm{K}.$$

These results are different from those given in Ref. 3, because the data of Refs. 7 and 9 were overlooked then.

The results of the fit are shown in Fig. 1 and all the data seem to be well represented by the fit. It should be pointed out that the data of Hoeve and van Ostenberg[7] are essential for the accurate determination of β.

The temperature scale thus determined from the macroscopic susceptibilities was then used to plot the local susceptibilities,[3,10,11] and the results are shown in Fig. 2. This figure also contains three straight lines which have been obtained in the following way. The dash-dotted curve is the result obtained in the previous analysis of the data by fitting a relation of the form $\chi_{loc} = a/(T+\theta)+b$ with

$$\chi_{loc} = ({}^{H}\mathrm{hf}/H_{ext})_{H_{ext}\to 0}\ .$$

In that fit all the data for $T \geq 60$ °K were used in order to be sure that any influence of a possible low-temperature anomaly was avoided. The fit

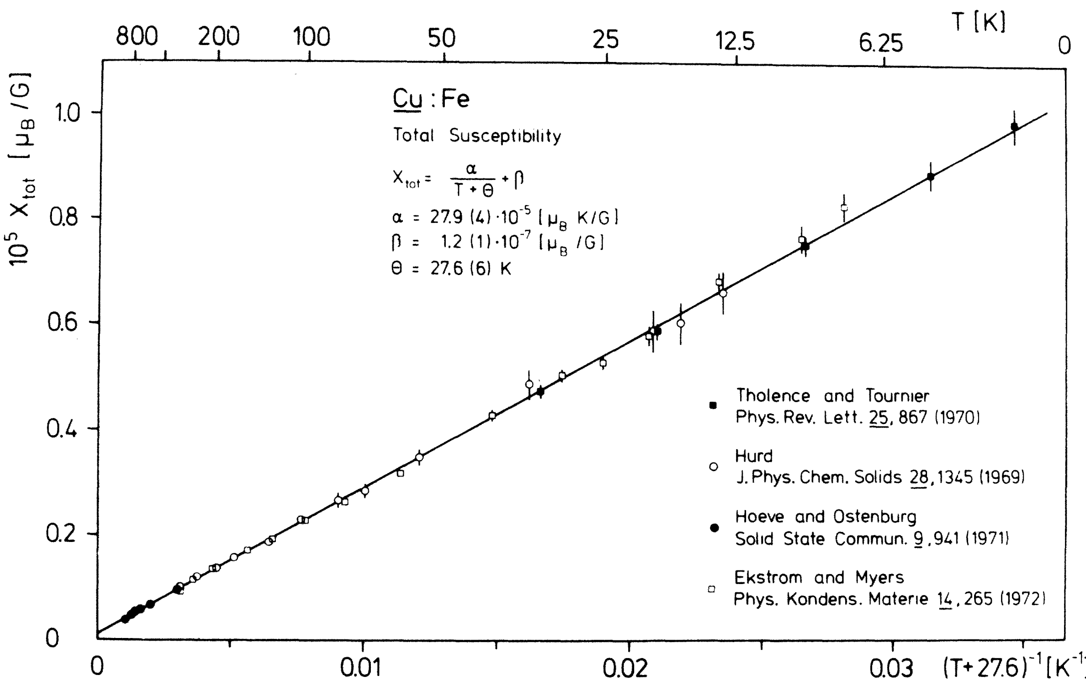

FIG. 1. Total susceptibility as a function of $1/(T+27.6)$ °K data are from Refs. 6–9. These data were fitted to a relation of the form $\chi_{tot} = \alpha/(T+\Theta) + \beta$ in order to obtain $\Theta = 27.6$ °K.

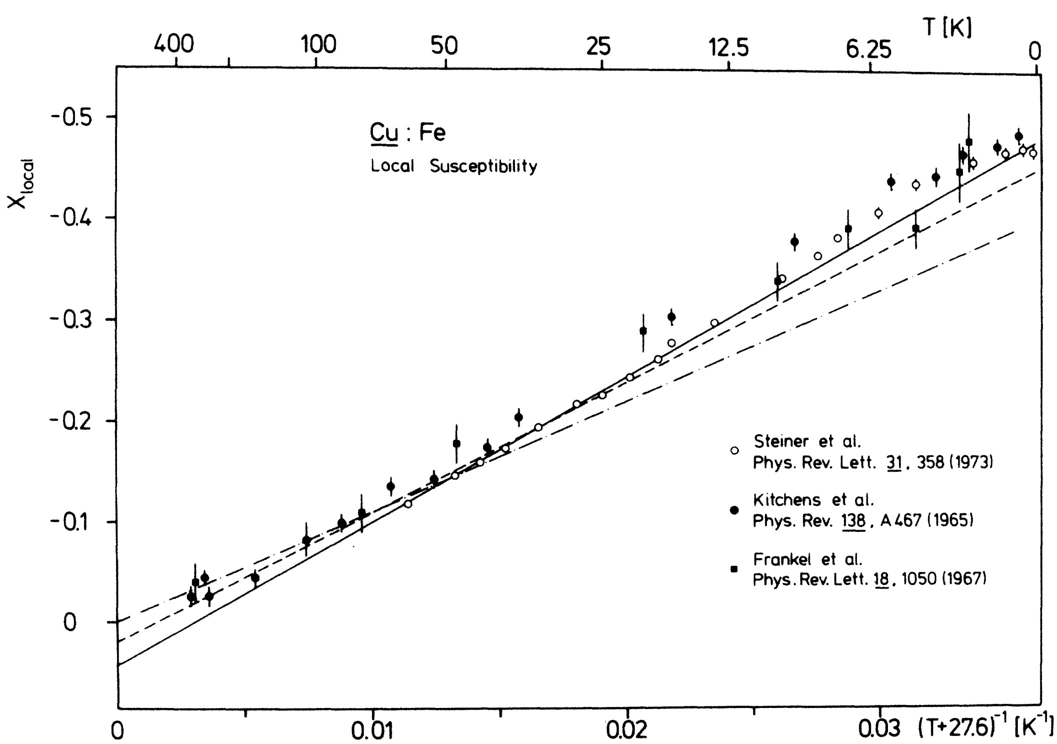

FIG. 2. Local susceptibility measured by Mössbauer experiments[3,10,11] as a function of $1/(T+27.6$ °K). Dash-dotted line fit from Ref. 3. Dashed line is a fit to the equation $\chi_{loc} = a/(T+27.6) + b$ using all data[3,10,11] for $T \geq 20$ °K; full curve uses all data for $T \geq 20$ °K from Ref. 3.

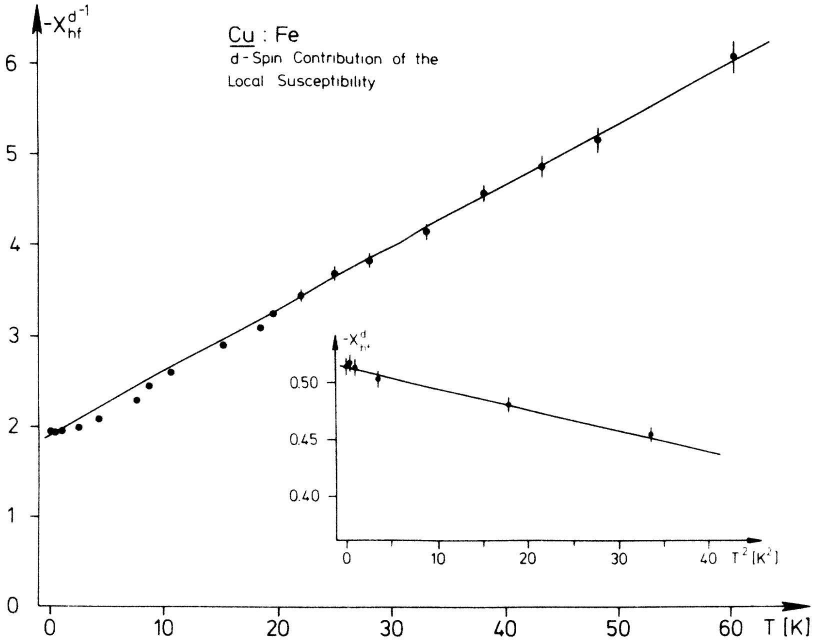

FIG. 3. Inverse local susceptibility corrected for the orbital contribution using the fit resulting in the full line in Fig. 2 as a function of T. The insert shows the low temperature part as a function of T^2, suggesting a T^2 behavior as first found by Schotte and Schotte[12] and Götze and Schlottmann.[13]

did not yield a value for b that was statistically significant. In the present analysis the range of data included in the fit was extended to lower temperatures until the quality of the fit decreased markedly; this occurred if data below 20 °K were included. The dashed curve was then obtained by fitting all hyperfine data above $T = 20$ °K to the above equation yielding $a = -12.9(6)$ and $b = 2.0(7) \times 10^{-2}$, which corresponds to an orbital contribution to the Knight shift of 2%. Finally, the full curve has been obtained by using only the data of Ref. 3 between 20 °K and 60 °K for the fit giving $a = -14.5(3)$ and $b = 4.5(4) \times 10^{-2}$. This essentially doubles the value for K_{orb} yielding $K_{orb} = 4.5\%$. The quality of the fit in this case is remarkably better than in the case discussed before using all existing data above 20 °K. Using the temperature-independent value of the total susceptibility $\beta = 1.2 \times 10^{-7}$ μ_B/G and an orbital hyperfine field of $H_{orb} = 500$ kG/μ_B (Ref. 5) for Fe, one would expect an orbital contribution to the hyperfine field of $K_{orb} = 6(1)\%$ which is quite close to the one obtained by fitting only the data of Ref. 3. But since at this point there is no reason to discard the data of Refs. 10 and 11 a definite value for K_{orb} cannot be given; clearly high-accuracy Mössbauer data in the temperature region between 100 °K and 300 °K could help to clarify this problem.

The analysis of the macroscopic and microscopic data suggests that there is indeed a temperature-independent orbital contribution to the susceptibilities as suggested in Refs. 4 and 5. But it is equally obvious from Fig. 2 that all the fits that treat the data in a statistically significant way do still lead to a low-temperature "anomaly" in the local susceptibility.

Finally in Fig. 3 the local d spin susceptibility is shown. This is the local susceptibility as measured by the Mössbauer data of Ref. 3 from which a constant orbital term as determined by the full line in Fig. 2 was subtracted. This susceptibility shows a slight nonlinear behavior at low temperatures and follows indeed the predicted T^2 dependence,[12,13] as can be seen from the insert. At this point it may perhaps be premature to decide whether the total susceptibility does not also show this type of temperature dependence. The only reliable data in this temperature region are those from Tholence and Tournier[8] and they are taken at rather large temperature steps, which makes the definite decision upon the power law a problem. This suggests that the final answer to that question can only come from further experiments.

The preceding analysis suggests that the hyperfine data as they stand cannot be fitted over the whole temperature range by a simple straight line. The full curve of Fig. 1 in Ref. 4 can only be a correct representation of the data if their errors are considerably larger than stated by the authors.[3,10,11] Therefore, the low-temperature "anomaly" in the susceptibility data stands, although it may be smaller than previously[3] assumed.

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