Theoretical explanation for the observed temperature dependence of the magnetic susceptibility of scandium

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Fermi-liquid theory is used to explain the observed maximum in the temperature dependence of the magnetic susceptibility of high-purity scandium. Previous calculations, based on Stoner theory and band calculations, are critically discussed. The observed disappearance of the maximum when small amounts of impurities are added is also explained.

The magnetic susceptibility χ of high-purity scandium was measured by Spedding and Croat[1]. A maximum in χ as a function of the temperature T was found at about 25 °K. The maximum was present only at the purest sample and it disappeared when small amounts of nonmagnetic impurities were added. An attempt to explain the maximum using Stoner theory and band calculations was recently reported by Das[2]. The purpose of this paper is to explain the experimental data, as well as the disappearance of the maximum due to impurities, on the basis of Fermi-liquid theory.

According to the Fermi-liquid theory for the temperature dependence of the susceptibility of normal paramagnetic metals[3–5], the existence of the interactions between the electrons as well as the presence of a sharp Fermi surface lead to the following nonanalytic temperature dependence of the susceptibility at low T:

$$\chi(T)=\chi(0)-bT^2\ln(T/T^*);\quad b>0. \tag{1}$$

This result is a consequence of the nonanalyticity of the self-energy Σ on the real frequency ω axis and therefore is not obtained in Stoner's theory, where the ω dependence of Σ is neglected.

Equation (1) leads to a maximum in $\chi(T)$ at $T=T^*/\sqrt{e}$ (higher-order terms may affect the actual position of the maximum). According to the "paramagnon model," the coefficient b is proportional to S^4(Ref. 5), where S is the Stoner enhancement factor. According to Misawa's calculations[6] $b\sim S^2$. In both cases, the effect is most likely to be observed in strongly enhanced ($S\gg 1$) paramagnetic metals and has in fact been observed in many such materials: Pd, α-Mn, U_2C_3, YCo_2, $LuCo_2$, $NpCo_2$, $CeSn_3$, Rh, Y, Pt, and others (see Refs. 4–7).

The maximum in $\chi(T)$ of scandium is therefore not a rare effect but rather one of a general type of behavior. The enhancement factor of Sc was estimated to be 4.6 (Ref. 2), which makes it a good candidate for the effect.

It is well known that the spin fluctuations play a most important role in strongly enhanced paramagnets. They are responsible, for instance, for the strong damping of the quasiparticles with energies close to the Fermi level. This effect is always attached to the Fermi level and leads to the strongly enhanced coefficient b.[5] Spin fluctuations are neglected in Stoner's model and therefore this model is not appropriate for the study of strongly enhanced materials. Spin fluctuation effects were included in the works of Beal-Monod *et al.*[8] and Kawabata.[9] They obtained a T^2 variation for $\chi(T)$ with a coefficient proportional to S^3 (Stoner theory gives S^2). However, in their diagrammatic treatment, they neglect self-energy insertions, the inclusion of which was shown[5] to lead to a complete cancellation of their T^2 term.

To check our theory in the case of Sc, we tried to fit the expression given by Eq. (1) to the experimental data of the susceptibility of the purest sample (Sc-4-155-B of Ref. 1) in the c direction, which exhibits the most pronounced maximum. We have found that the formula

$$\chi(T)=8.2-(T/52.3)^2\ln(T/43.7) \tag{2}$$

(in 10^{-6} emu/g) fits the observed data in the temperature range between 6 °K and 40 °K with a rms% deviation of less than 0.2% (Fig. 1). Above 35 °K, the higher-order terms of $\chi(T)$ are important and the experimental data may be fitted over a wider range of temperature by introducing a term proportional to $T^3\ln T$(Ref. 4) or $T^4\ln T$.[3,6]

The increase of $\chi(T)$ below 6 °K is presumed to come from magnetic impurities.[1]

Attempts have been made to calculate $\chi(T)$ in Pd(Ref. 10), in Sc[2], and in Pt(Ref. 11) using Stoner theory and band-structure calculations, but these calculations are limited by the histogram size of the calculated density-of-states curve. Since the relevant part of the density-of-states lies in a region of the order kT around the Fermi level, one must know the accurate density of states in this region. Although one can always use extrapolated density of states by polynomial

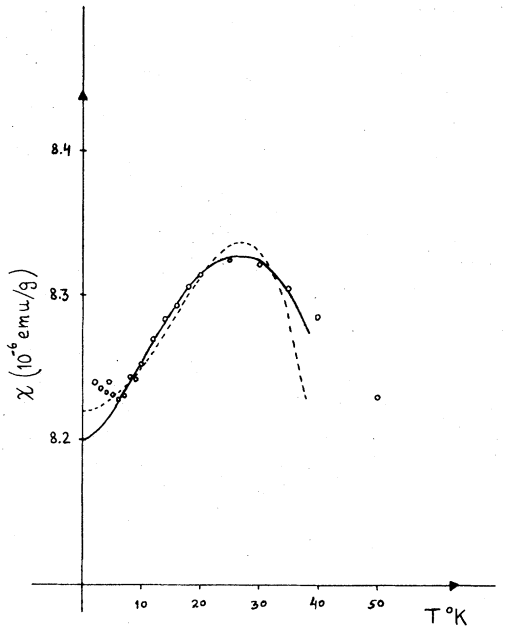

FIG. 1. Temperature dependence of the susceptibility of Sc. The experimental points are the results of Spedding and Croat (Ref. 1) (sample 4-155-B, χ_c). The bold theoretical curve given is Eq. (2). The broken line is the best fit using a three-parameter Stoner theory.

fit, this procedure certainly obscurs any important changes in the derivatives of the density of states close to the Fermi level. These derivatives are vital in any calculation of the temperature dependence of the susceptibility. In palladium, the accuracy of band calculations is limited to 1 mRy (Ref. 12) (~ 160 °K), while the maximum in $\chi(T)$ occurs at approximately 85 °K. In scandium, the accuracy is limited to a few millirydbergs, [2] while the maximum occurs at approximately 25 °K. In platinum[11] band-structure density of states were calculated with an energy mesh of 0.25 mRy. Their calculated $\chi(T)$ agreed well with the experimental data of Budworth *et al.*[13] between 100 and 220 °K. In this case, no maximum in $\chi(T)$ were obtained neither in the experiment nor in the calculations. However, later measurements by Foner *et al.*[14] show a small maximum at 100 °K. Thus band-structure calculations do not reproduce the new measured data while Fermi-liquid theory (see Ref. 7) agrees perfectly well with measurement in the temperature range between 30 and 190 °K.

In Stoner's theory, the position of the maximum in $\chi(T)$ and its height above $\chi(0)$ depend strongly on the delicate structure in the density of states around the Fermi level[15]. That explains why, in using band calculations, the position of the peak in $\chi(T)$ was found to be very sensitive to the position of the Fermi level and indeed, at least in the case of Pd (Ref 10) the position of the Fermi level was used as an extra adjustable parameter. In this way, a maximum in $\chi(T)$ was obtained, but the overall fit to the experimental data is poor. In the case of scandium,[2] a sharp dent in $\chi(T)$ was obtained instead of the observed smooth maximum.

Even if the density of states was known with the required accuracy, Stoner's theory still predicts the following analytic expansion of the susceptibility for low T:

$$\chi(T) = \chi(0) + \sum_n a_n T^{2n} , \qquad (3)$$

where the coefficients a_n depend on the density of states and its derivatives with respect to the energy, evaluated at the Fermi level.

The maximum in $\chi(T)$ is then attributed to special feature of the bare density of states near the Fermi level, [15, 16] but since similar maxima showing a characteristic $T^2 \ln T$ behavior appear in so many different strongly enhanced paramagnetic metals and metallic compounds, one would have to postulate the existence of these special features in the bare density of states in all these materials, always of the same form and always situated precisely at the Fermi level. According to the Fermi-liquid theory, the nonanalyticity of the self-energy is always at the Fermi energy and therefore it provides a natural explanation to the widely observed effect. Furthermore, Stoner's theory does not exclude the possibility of any other temperature variation of χ, say the existence of a minimum. Experimentally, a maximum is observed in *all* strongly enhanced pure paramagnetic metals and metallic compounds.

The coefficient b in Eq.(1) has been calculated within the paramagnon model approximation[5] and it agreed well with the coefficient obtained by least-mean-square fitting in the case of Pd, with $S \sim 10$. This did not work in the case of Sc, presumably because S is not large enough and because compared with Pd, the Fermi level is too far away from the edge of the band[2] for the paramagnon model to be valid. The parameter T^* in Eq. (1) is roughly proportional to the spin-fluctuation temperature, i.e., T_F/S, where T_F is the Fermi temperature, but since this parameter also absorbs in it the coefficient of the regular T^2 term in χ arising from the band contribution and regular terms in the self-energy, it is so far not possible to calculate it theoretically. T^* remains a parameter to be determined by experiment.

Disregarding the paramagnon model value for b,

Eq. (1) consists of a three-parameter theory to fit the experimental data. To compare this with a three-parameter Stoner theory, we tried to fit the experimental data to a variation of the form $\chi = \chi(0) + aT^2 + bT^4$. The best fit using the least-mean-square method is shown in Fig. 1 (broken line), thus exhibiting the supremacy of the nonanalytic variation given by (1) over the analytic expansion (3).

The effects of impurities on the logarithmic term have been studied in a previous work.[5] Using a paramagnon model calculation in the case of a finite mean free path, [17] the following low-temperature variation of χ was obtained:

$$\chi(T) = \chi(0) - bT^2 \ln[(T + T_{\text{imp}})/T^*] , \qquad (4)$$

where

$$T_{\text{imp}} = T_F (S k_F l)^{-1} .$$

k_F is the Fermi momentum and l the mean free path.

The maximum measured from $\chi(0)$ is then reduced in magnitude and its position shifted to lower temperatures. The maximum disappears completely when T_{imp} reaches the value of T^*. This behavior is observed in palladium alloys such as *Pd*–Ag(Ref. 18), *Pd*–Rh(Ref. 19), and *Pd*–Pt (Ref. 20) and it explains the disappearance of the maximum in Sc when small amounts of impurities are added to the purest sample.[1]

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