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Analysis of the CESR ferromagnetic and paramagnetic itinerant electron compounds: ZrZn_2 , Sc_3In vs TiBe_2

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(14. XII. 1987)

In honor of Martin Peter's 60th birthday.

Abstract. Analysis of the conduction electron spin resonance CESR in the weak itinerant ferromagnets ZrZn_2 and Sc_3In in their paramagnetic state gives different relaxation behavior as compared to the weak itinerant paramagnet TiBe_2 . In the ferromagnetic compounds the increase of the line width with temperature can be related mostly to the variation of the enhancement factor but in TiBe_2 to the change in the resistivity. It is suggested that the CESR of TiBe_2 is due to d electrons in pockets at the Fermi surface. These d electrons are hybridized with s electrons at low temperatures and dehybridized above 200 K. This assumption is supported by the temperature behavior of the line width in the $\text{Ti}(\text{Be}_{1-x}\text{Cu}_x)_2$ system whose susceptibility increases with x and becomes ferromagnetic for $x > 0.07$.

Introduction

Among the weak itinerant magnetic metals the intermetallic compounds ZrZn_2 , Sc_3In and TiBe_2 do not contain magnetic elements such as Fe, Co and Ni. At high temperatures their susceptibility has a Curie Weiss behavior [1, 2, 3]. At low temperatures the first two compounds are ferromagnetic [1, 2] while TiBe_2 remains paramagnetic and has a maximum in its susceptibility around 10 K [4]. These compounds are of interest (besides being magnetic without a magnetic element) as their elements cannot form, due to disorder, magnetic clusters. Such clusters can interfere with the investigation of the properties at low temperatures, and can give erroneous interpretation or doubts on their basic properties.

Their magnetic properties are attributed to the band structure and the large density of states at the Fermi surface [5]. It is possible to vary these properties such as the Curie temperature by substituting part of their elements with other elements [6, 7]. TiBe_2 is of particular interest. It is paramagnetic but its susceptibility is not linear with the magnetic field and has a maximum around 5T [8]. When substituting 7% of the Be with Cu it becomes ferromagnetic and with 50% Cu the Curie temperature is about 33 K [7]. The investigation of these

materials was focused mostly on their low temperature properties as it was expected that they would enable to obtain a better understanding of the mechanism of interaction between the electrons that give rise to the magnetic properties [9]. However, the theoretical interpretation of the low temperature experimental results depends on the very detailed structure of the density of states (DOS). Though very detailed calculations of the DOS were performed in these compounds, and in particular in TiBe_2 [10], so far they are not fine enough. A complementary approach suggested by Polatzek and Zevin [11] is to calculate the properties at high temperatures where the detailed structure of the DOS is less important.

In this work we would like to discuss the experimental results of the conduction electron spin resonance (CESR) of these compounds at elevated temperatures in their paramagnetic states in conjunction with the above remark. As will be shown one can obtain from the high temperature CESR results an insight on the nature of interactions in these interesting materials.

Experimental

The magnetic resonance was observed in ZrZn_2 [12] and Sc_3In [13] in the ferromagnetic as well as in the paramagnetic state. The line widths as a function of temperature is shown in Fig. 1 (taken from Refs. 12 and 13). The low

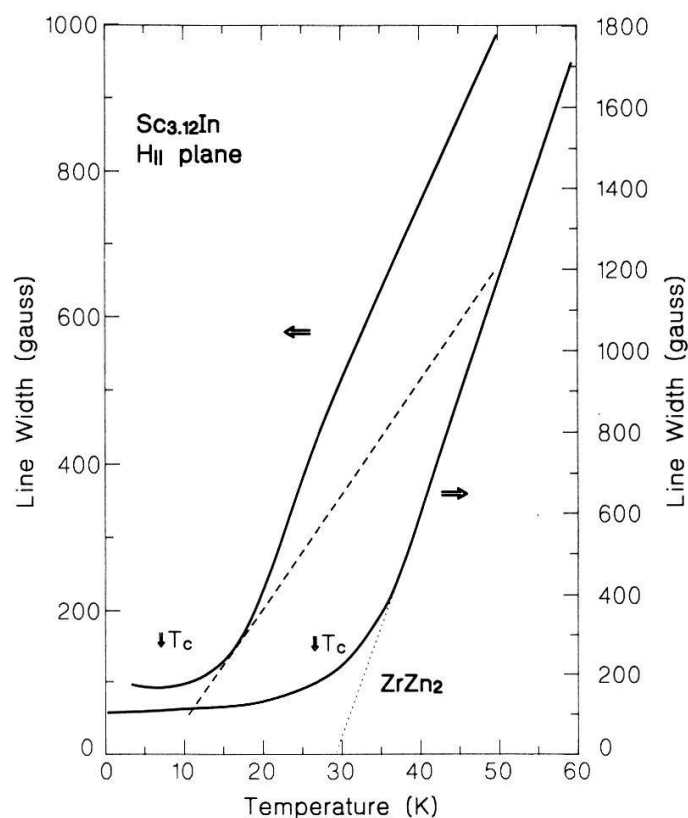


Figure 1

Line width as a function of temperature in ZrZn_2 , Sc_3In (taken from Ref. 12 and 13 respectively). The broken lines are a fit to $1/M$ behavior.

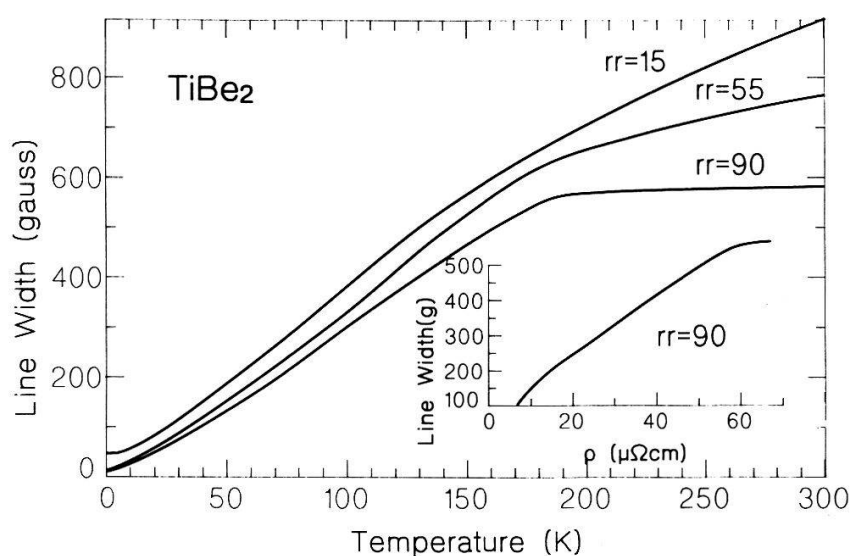


Figure 2

Line width as a function of temperature in TiBe_2 (Ref. 15). Line width as resistivity in TiBe_2 where the temperature is the implicit parameter for a resistivity ratio of 110 (Ref. 14)

temperature part that contains the shallow minimum is the ferromagnetic resonance and will not be discussed here. Above the Curie temperature, in the paramagnetic state, a very sharp increase of the line width with temperature is observed, with a slope of 40 and 25 gauss/K for ZrZn_2 and Sc_3In respectively. These results should be compared with the CESR of the paramagnetic TiBe_2 compound [14] where an increase of 3 gauss/K is observed. (Figure 2 taken from Ref. 14). This relatively small increase enabled the observation of the resonance up to room temperature. Above about 200 K the slope decreases and it depends on the resistivity ratio of the sample [15]. This peculiar and interesting behavior at high temperature will be discussed further below. The g value in the paramagnetic state of ZrZn_2 [12] and Sc_3In [13] was reported to be 1.98 and 1.99 respectively. For TiBe_2 a g value of 1.983 was reported [15].

Theoretical considerations

We will first discuss the proposed mechanism for the temperature behavior of the line width. It is assumed that the line width is mostly due to the spin lattice relaxation time $T_1 = T_2$, and is caused by spin orbit coupling transport collisions. In other words for each momentum scattering there is a probability of spin flip that increases with the strength of the spin orbit coupling [16]. The relaxation time T_1 is given by the Elliot relation [16] $T_1 = \tau/(\Delta g)^2$ where τ is the momentum scattering time and Δg is the g shift away from the free electron g value of 2.00.

Numerical evaluation in ZrZn_2 and Sc_3In where τ is obtained from resistivity measurements [12, 13] gives a line width of at least 50 times larger than the experimental results at temperatures slightly above the Curie temperature. For TiBe_2 the calculated line width is about 120 times larger (e.g. 2400 gauss) compared to 20 gauss observed at 1.4 K for a sample with a resistivity ratio (rr) of

110 [14]. However, microscopic calculations predict [17] that the relaxation rate in exchange enhanced system is reduced by the enhancement factor $1/(1 + B_0) = 1 + \lambda\chi$ where B_0 and λ are the enhancement parameter of a Fermi liquid and the exchange constant respectively.

Walsh et al [12] have applied this narrowing argument for ZrZn_2 in the paramagnetic state by using an equivalent assumption that the spin relaxation is towards the instantaneous field and therefore the relaxation time is enhanced by the enhancement factor $1 + \lambda M/H$. This reduces the line width of ZrZn_2 by a factor of 40 slightly above the Curie temperature in good agreement with the experimental results. Furthermore, the temperature dependence of the line width is also explained by this approach as $M/(T) = 1/(T - \theta)$ and therefore $\Delta H \propto (T - \theta)$ as shown in Fig. 1. The same argument holds for Sc_3In although the quantitative agreement is smaller [13] (Fig. 1). In these two compounds the variation T_1 as a function of temperature was not taken into account. But as the line width measurement in the paramagnetic state was observed, due to the large broadening, only in a small temperature range, the corrections introduced from the resistivity measurements are small compared to the variation of the line width due to the change in the magnetization, and could therefore change only slightly the numerical results.

For TiBe_2 where a line width of less than 20 gauss is observed for $rr = 110$ at 4.2 K, the narrowing argument in exchanged enhanced system reduces the lines width by a factor of about 100 in agreement with experimental results. However, contrary to the ferromagnetic compounds the increase of the line width with temperature up to 200 K can be obtained by assuming that the variation of the line width is only due to the variation of τ in the Elliot relation [14], and consequently the enhancement factor that is responsible for the narrow line width at low temperature remains constant even at high temperatures. The variation of τ with temperatures is obtained from the variation of ρ and as seen from Fig. 2 the variation of the line width with the resistivity is linear up to about 200 K in agreement with the above considerations.

Above 200 K the increase of the line width with the resistivity depends on the resistivity ratio and for the highest rr sample of 110 there seems to be a saturation where the line width remains constant with increasing resistivity and temperature. As explained elsewhere [15] this saturation may arise from dehybridization of s and d electrons which affects the resistivity behavior at high temperatures. This dehybridization approach was suggested recently by Weger and Mott [18]. At low temperatures the s and d electrons are hybridized via the $s - d$ hybridization integral J_{sd} . However, due to the decrease of the relaxation time with temperature the s and d electrons dehybridized and they contribute separately to the resistivity. However, due to the Ioffe–Regel rule [19], above a certain temperature the resistivity of the d electrons saturates and therefore they cannot add to the increase of the resistivity with temperature, but the s electrons continue to contribute to this increase. Thus, for high rr TiBe_2 and above about 200 K the line width remains constant indicating that the resonance observed is due to d electrons. The effect of the rr on the high temperature behavior of the line width will be discussed elsewhere [15].

Discussion

The comparison of the spin relaxation in the paramagnetic weak itinerant metal and in the ferromagnetic weak itinerant metals in its paramagnetic state shows different behavior. In the ZrZn_2 and Sc_3In the increase in line width can be obtained by assuming the decrease of the enhancement factor which is proportional to the M/H . In contrast, in TiBe_2 the spin relaxation can be directly related to the momentum relaxation and is not affected by the variation of $\chi = M/H$ for the temperature range 0–300 K. The saturation of the resonance line width above about 200 K in TiBe_2 indicates that the electron spin resonance originates from d electrons. Thus not all the electrons at the Fermi surface participate in the observed electron spin resonance. Jalborg et al [10] have calculated the Fermi surface structure. They found that the largest contribution to the density of states are Ti d electrons. It is reasonable to assume that the resonance observed are due to Ti d electrons. However, as the susceptibility and therefore the enhancement factor decreases with the increase of temperature, an additional increase of the line width with temperature is expected as it was found in the ferromagnetic compounds, (i.e. as the susceptibility decreases by a factor of 6 and 9 from helium to 200 K and room temperature respectively, this should increase the line width by the same factors). This additional contribution was not observed.

A way out of this contradiction is to assume that in TiBe_2 the resonance of the observed d electrons are due to d electron pockets in the Fermi surface, whose enhancement factor remains constant with temperature even at elevated temperatures, and that their contribution to the total magnetization is small. In the weak ferromagnetic itinerant metals the interaction between the various electrons in the Fermi surface is much stronger and therefore they will respond to the total magnetization as observed in ZrZn_2 and Sc_3In . A support to this can be obtained from the EPR studies of $\text{Ti}(\text{Be}_{1-x}\text{Cu}_x)_2$ in their paramagnetic state. As mentioned earlier for $x > 0.07$ the compound becomes ferromagnetic and the Curie temperature increases with x . The magnetic resonance shows a smooth transition from the paramagnetic compound to the ferromagnetic compounds which is obtained by increasing x . A simple criteria for the strength of the interaction can be obtained from the average slope of the line width with temperature. As shown in Fig. 3 (taken from Ref. 14), the slope increases with x from $3\text{g}/^\circ\text{K}$ to $15\text{g}/^\circ\text{K}$ for $x = 0$ and 0.2 respectively. For $x = 0.2$ the Curie temperature is 18 K and the general features of the line width vs temperature are very similar to those of ZrZn_2 and Sc_3In shown in Fig. 1.

In conclusion, this work shows a different spin relaxation behavior in weak itinerant paramagnetic and ferromagnetic compounds as observed via their electron spin resonance in their paramagnetic state. This difference may arise from the strength of the interaction between the different conduction electrons. It calls for a theory on the temperature dependence of the enhancement factor based on the particular structure of the Fermi surface and taking into account electron correlation. The ESR results in $\text{Ti}(\text{Be}_{1-x}\text{Cu}_x)_2$ show that these compounds present a system where there is a continuous variation of the spin

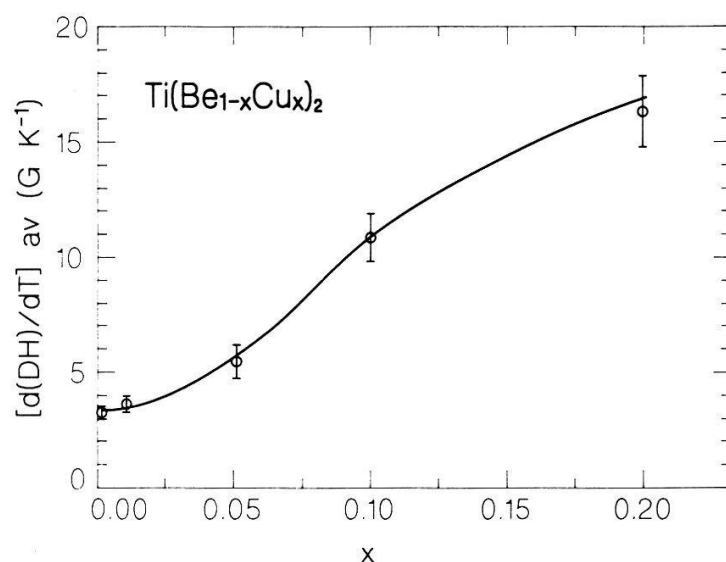


Figure 3
The average slope $d(DH)/dt$ as a function of x in $Ti(Be_{1-x}Cu_x)_2$ (Ref. 14)

behavior from the paramagnetic to the ferromagnetic state. A systematic study of this system where magnetization, resistivity, ESR and other properties performed on the same samples, may contribute to a better understanding of the behavior of the conduction electron in weak itinerant metals and the transition from an itinerant paramagnetic to an itinerant ferromagnetic compound.

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