Paramagnetic Resonance in the Cubic Helimagnet \( \epsilon \)-FeGe

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Abstract. We have investigated the spin dynamics of the paramagnetic region of \( \epsilon \)-FeGe by means of Electron Spin Resonance (ESR) spectroscopy. The high-quality single crystal showed a strong paramagnetic resonance line which is well-described by a Lorentzian shape above 287 K. The close relation of the linewidth to the magnetic susceptibility indicates a resonance of a strongly coupled 3d / conduction electron system.

1. Introduction

Investigations of ferromagnetic metals by the Electron Spin Resonance (ESR) technique provide valuable information on the spin dynamics in the paramagnetic regime. Important examples of 3d metals are TiBe\(_2\) [1], ZrZn\(_2\), NbFe\(_2\) [2], and MnSi [3, 4] which allow an ESR investigation of the paramagnetic resonance without doping of ESR probe spins and at temperatures low enough to be easily accessible by the ESR technique (unlike the temperatures needed for Fe and Ni paramagnetic resonances [5]). They all have in common an ESR linewidth that is mainly determined by ferromagnetic correlations which considerably enhance the paramagnetic susceptibility. The observation of such correlation effects in ESR has been found to be relevant also for 4f heavy fermion metals [6, 7] and allows considerable insight into the nature of metallic magnetism.

Detailed magnetic measurements on the cubic helimagnet \( \epsilon \)-FeGe (space group \( P2_13 \)) revealed for temperatures below \( T_C = 278.2 \) K a rich variety of complex magnetically ordered phases as well as field dependent precursor phenomena above \( T_C \) [8]. Here, we report ESR investigations in the paramagnetic regime providing one additional example for the ESR observability of paramagnetic resonances in ferromagnetic metals without the need of doping ESR-active centers in the material.
Figure 1. Four representative resonance lines at 9.4 GHz (X-band) for temperatures above magnetic ordering. Red lines show a Lorentzian shape of a paramagnetic resonance which provides an accurate spectra fitting for $T \gtrsim 287$ K only. Amplitudes were normalized for better illustration. Inset displays the resonance field $H_r$ as a function of rotation angle around the [110] axis ($\perp \bf{H}$). Solid line shows uniaxial fit.

2. Experiment and Results
We measured the power $P$ of a magnetic microwave field $\bf{h}$ absorbed by an high-quality $\epsilon$-FeGe single crystal [8] as a function of a transverse external static magnetic field $\bf{H}$ ($\bf{H} \perp \bf{h}$). The crystal was mounted with its [110] axis nearly parallel to the $\bf{h}$-field which results in a $\approx 45^\circ$ orientation of its [100] axis to the fields $\bf{H}$ and $\bf{h}$. The temperature was varied with a $^4$He-flow-type cryostat. The paramagnetic resonance was recorded as the derivative $dP/dH$ because a lock-in technique was used to enhance the signal-to-noise ratio. The parameters of the line (resonance field $H_r$, linewidth $\Delta H$, intensity $I^{\text{ESR}}$) were evaluated at three microwave frequencies $\omega/2\pi = 1.1, 9.4, 34$ GHz (L-, X-, Q-band). They contain the static and dynamic magnetic properties of the resonating spin system which is characterized by the $g$-factor $g^{\text{ESR}} \propto \omega/H_{\text{res}}$, the relaxation rate $\Gamma \propto \Delta H$ and the intensity $I^{\text{ESR}} \propto \chi_0$ (static magnetic susceptibility).

Figure 1 shows four examples of well-defined spectra of $\epsilon$-FeGe for three temperatures above $T_C$. The spectra consist of strong resonance absorptions without any traces of weak and narrow lines which would indicate spurious Fe-containing phases. The lineshape could be well described by a metallic Lorentzian [9] for $T \gtrsim 287$ K. In the framework of a conduction electron spin resonance this metallic Lorentzian would correspond to a Dysonian lineshape [10] in the limit of a much faster relaxation of a conduction electron spin than its diffusion through the skin depth. Hence, from the shape, one cannot distinguish whether the resonance in $\epsilon$-FeGe arises from a local or an itinerant spin probe. This distinction is possible, for instance, for the paramagnetic line of Fe$_{0.5}$Co$_{0.5}$Si for which a Dysonian lineshape with a much faster spin-diffusion than spin-relaxation was used [11].

Below $T \approx 287$ K deviations from a Lorentzian shape as well as from a Dysonian shape
Figure 2. Left: Temperature dependence of ESR intensity and comparison with the static dc-susceptibility $\chi_0$ at $\mu_0 H = 0.32$ T (inset). Right: ESR $g$ factor and linewidth $\Delta H$ for L-, X-, and Q-band frequencies. The reciprocal ac-susceptibility $\chi_{ac}^{-1}$ (right axis) was measured at $0.225 T$ [8], close to the X-band resonance field (0.32 T). Solid lines emphasize a linear behavior as discussed in the main text.

gradually appear (compare red lines with data) and become pronounced below $\approx 284$ K. These shape distortions are unusual for a purely paramagnetic resonance and occur in the same temperature region where the ac-susceptibility shows a maximum and several crossover lines have been found in the phase diagram above $T_C$ [8]. In this precursor region short-range magnetic modulations build up which could be responsible for the observed line shape distortions. Upon entering the region of magnetic ordering a variety of ferromagnetic resonance modes appear at fields very similar to those reported earlier from polycrystalline FeGe [12]. Similar features where also reported for the resonance modes in the ordered states of MnSi and Fe$_x$Co$_{1-x}$Si where a thorough investigation regarding lineshape and resonance field was performed [3, 4, 13, 14, 15]. These results might be applicable for $\epsilon$-FeGe as well. In this paper, however, we refrain from presenting further details of the resonance in the ordered region of $\epsilon$-FeGe and focus on the resonance in the paramagnetic regime instead.

Upon rotating the crystal around its [110] axis (which is aligned $\perp H$) the resonance field varies with a 180°-periodicity as shown in the inset of Fig.1 for $T = 294$ K. A uniaxial fit (solid line) corresponds to $g_{45^\circ} = 2.107 \pm 0.004$ and $g_{135^\circ} = 2.082 \pm 0.004$. However, these $g$ factors may not reflect the anisotropy caused by off-diagonal crystalline electric field parameters in a cubic crystal symmetry because then a structure with a 90°-periodicity would be likely for the applied rotation geometry [16]. Moreover, the amplitude of the resonance field variation decreases by 50% with increasing the temperature from 290 K to 300 K. Therefore, despite the almost spherical shape of the crystal, we suspect very weak demagnetization fields to contribute to the resonance field.

Figure 2 shows the temperature dependence of various resonance line parameters. The ESR intensity, obtained from doubly integrating the $dP/dH$-spectra, measures the static magnetic susceptibility of the resonating probe. This could nicely be confirmed with the linearity between the ESR intensity and the dc-susceptibility $\chi_0$ shown for X-band in the inset.
The ESR $g$-factor and ESR linewidth $\Delta H$ were both obtained from Lorentzian line-fittings, keeping the dispersion-absorption ratio constant. The ESR $g$-factor shows a temperature dependence which is strongest for the smallest resonance field (i.e. for the L-band measurements). This behavior indicates the direct relation between the field dependent dc-susceptibility and the ESR $g$-factor which, hence, measures the magnetic molecular-field. The linewidth displays a reciprocal dependence to the ac-susceptibility, $\chi_{ac}$. This behavior is a hallmark of a resonance of a strongly coupled 3d / conduction electron system [17]. Therefore, the linear behavior of $\Delta H$ with a slope $b = 5$ mT/K as shown by the solid lines is not indicating a Korringa relaxation mechanism as is often the case for the relaxation of diluted ESR probes in a metallic environment [17].

While a frequency dependence of the resonance for $T > 290$K is not visible in the $g$ factor the linewidth shows a linear broadening with $0.8 \pm 0.1$ mT/GHz. This behavior is in agreement with the results for Fe$_{1-x}$Co$_x$Si ($\approx 1.2$ mT/GHz) [11] but in contrast to the frequency independence of the linewidth in systems like ZrZn$_2$ [18]. There, the broadening mechanism is solely determined by the exchange enhanced relaxation of the conduction electron spins [19]. Hence, the frequency dependence of the linewidth in $\epsilon$-FeGe indicates additional relaxation mechanisms.

3. Conclusion

In the paramagnetic regime of $\epsilon$-FeGe, above temperatures of $\approx 287$ K, the spin dynamics of the strongly coupled Fe-3d / conduction electron system can be characterized by a field-dependent, almost isotropic paramagnetic resonance. The deviations from a Lorentzian line shape observed for $T_C < T < 287$ K are ascribed to complex magnetic short-range modulations which evolve in this precursor region.

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