Magnetoelastic quantum oscillations in GdSb to 55 T

R Daou\textsuperscript{1,2}, A Haase\textsuperscript{1,2}, M Doerr\textsuperscript{3}, M Rotter\textsuperscript{4}, F Weickert\textsuperscript{1,2}, M Nicklas\textsuperscript{1} and F Steglich\textsuperscript{1}

\textsuperscript{1} Max Planck Institute for Chemical Physics of Solids, D-01187 Dresden, Germany.
\textsuperscript{2} Hochfeld-Magnetlabor Dresden, FZ Dresden-Rossendorf e.V., D-01314 Dresden, Germany.
\textsuperscript{3} Technische Universität Dresden, Institut für Festkörperphysik, D-01062 Dresden, Germany.
\textsuperscript{4} University of Oxford, Dept. of Physics, Clarendon Lab., Parks Road, OX1 3PU Oxford, UK.

E-mail: Ramzy.Daou@cpfs.mpg.de

Abstract. The rare-earth monopnictide materials include many examples of strongly correlated electron materials with low carrier density and magnetic ground states. The semimetal GdSb orders antiferromagnetically below 23.4 K, and at low temperature a fully polarised ferromagnetic state is reached for applied magnetic fields greater than the saturation field, $H_s$, of 33.5 T. Magnetostriction measurements in pulsed magnetic fields up to 55 T show that the magnetoelastic coupling to the Gd$^{3+}$ moment system changes sign on crossing $H_s$. We also observe large-amplitude quantum oscillations of the sample length.

1. Introduction

The Gd monopnictides GdX ($X = N, P, As, Sb, Bi$) have attracted particular attention [1, 2] because of the ground state and fundamental magnetic interactions can be studied easily. All of them adopt the f.c.c. NaCl structure. Since Gd$^{3+}$ with $S = 7/2$ has no orbital angular momentum, they are pure spin systems and therefore no single-ion anisotropy is expected. Most of the magnetic properties can be explained by a Heisenberg molecular field model. The two-ion coupling leads to antiferromagnetic ordering with a Néel temperature, $T_N$, of 23.4 K for GdSb [2]. The standard model of rare-earth magnetism predicts that the magnetic field required to fully align the Gd$^{3+}$ moments, $H_s = \frac{3k_B(T_N-\theta_W)}{g_J\mu_B(J+1)}$ where $k_B$ is the Boltzmann constant, $\theta_W = -31.3 \text{ K}$ [3] is the Weiss temperature, $\mu_B$ is the Bohr magneton and $g_J = 2$ is the $g$-factor. From these values we predict $H_s = 27 \text{ T}$, which is close to the observed value of 33.5 T.

The progression of magnetic phases is first from a simple antiferromagnetic (AFM) ground state into a spin-flop state at low applied field (~ 0.5 T). The canting angle between spins is then smoothly decreased as the field is increased until a critical value, $H_c$, is reached. At higher fields the response is paramagnetic (PM). Thus at intermediate temperatures $T < T_N$, $H_c < H_s$.

The electrical transport properties of the Gd-monopnictides are also of interest as they are examples of clean, nearly compensated low carrier-concentration magnetic materials. While the rest of the series are antiferromagnetic semimetals, GdN is found to be a half-metallic, ferromagnetic semiconductor [4] with potential applications in spintronics. Bandstructure calculations [5, 6] find semimetallic ground states and can reproduce the increasing trend of $H_s$ with anion mass by incorporating small nearest- and next-nearest-neighbour interactions. Because of the high saturation field, GdSb has not been well investigated experimentally. Previous measurements of magnetization and magnetostriction [3, 7] in GdSb did not determine...
Figure 1. The linear magnetostriction of GdSb at various temperatures, relative to the value at 2.1 K and 0 T. The curves are offset by the relative thermal expansion of the sample. The data shown was taken during the upsweep of the pulsed magnetic field; the downsweeps match well. Inset: The critical field $H_c$ as a function of temperature.

2. Methods

The single crystal of GdSb used in this study was prepared at AMES laboratory, Iowa (ED088/3). The crystal dimensions along principal axes were $1.4 \times 1.6 \times 3.0$ mm$^3$ and its mass was 23 mg.

Measurements of the longitudinal magnetostriction were performed using a fibre optic strain gauge glued to the sample with cyanoacrylate epoxy. Details of the method can be found in Ref. [8]. This technique is suitable for the challenging environment of pulsed magnetic fields, and it offers a resolution for the relative length change of the sample, $\Delta L/L$, of about $10^{-7}$ ($\Delta L \sim 3$ Å for the current sample). While superconducting magnets allow better measurements, this is the highest resolution so far achieved in pulsed fields. The capacitor-driven pulsed magnet was energised to a maximum field of 55 T in a rise time of 33 ms. All of the data shown is taken from the upsweeps, but the data from the downsweeps match very well.

3. Results

In Figure 1 we show the linear magnetostriction of GdSb at several temperatures between 2.1 K and 26 K. Both magnetic field and the measured elongation were aligned along a principal axis of the crystal. Below 1 T the spin flop transition can be seen as a rapid increase in sample length.
In the spin-flop phase the sample length decreases smoothly as the moments rotate to align with the field. The critical field \( H_c \) is sharply defined by a kink in the data at high fields for \( T < T_N \), and the inset to Figure 1 shows the magnetic phase diagram described by these points. In the paramagnetic phase above \( T_N \), the sample length increases monotonically with field. At high fields and low temperatures, quantum oscillations in the sample length can be observed.

The relative thermal expansion is negative in the antiferromagnetic and spin-flop phases, but positive in the polarised paramagnetic phase. This can be explained by a change in sign of the strain dependence of the exchange interaction between antiferro- and ferromagnetic configurations, as expected, for example, for the RKKY mechanism.

In Figure 2 we have subtracted the background magnetostriction to focus on the quantum oscillations in the strain using fourth order polynomial fits for the low- and high-field data that are constrained to meet at \( H_c \). Below \( H_c \), only one quantum oscillation frequency can be resolved. However, above \( H_c \) there is a clear change in the periodicity of the oscillations that cannot be well fitted by a single frequency.

Bandstructure calculations predict the presence of several small ellipsoidal Fermi surface sheets, with electron pockets at the \( X \)-point and hole pockets at the \( \Gamma \)-point\[5, 6\]. The exact number and size of these pockets is somewhat sensitive to the position of the Fermi level. A calculation in the ferromagnetic state \[9\] shows that strong spin-splitting is expected in all sheets. The expected doubling of the unit cell size to reflect the periodicity of magnetic order in the AFM state also has consequences for the electronic structure \[10\]. The pockets at the \( \Gamma \)-point are so small, however, that they are not expected to be changed by the unfolding of the bands.

The temperature dependence of the amplitude of the observed frequencies is reasonably consistent with the Lifshitz-Kosevich formula \[11\], resulting in effective masses in the range 0.3–0.6 \( m_e \), similar to those reported in Ref. \[2\]. We also estimate the Dingle temperature for the \( F_0 \) frequency to be \((17 \pm 3) \) K from the field dependence of the 2.1 K data for \( H < H_c \). The Dingle temperatures for \( F_1−4 \) are harder to extract as there are only a few periods.

There are two possible explanations for the change in the frequency spectrum across \( H_c \). In the first scenario, the onset of AFM order causes a reconstruction of the Fermi surface, as has been observed in \( \text{NdB}_6 \)[10]. In the second, the Fermi surface remains unchanged through the AFM-PM transition but the frequency spectrum becomes more complex as a result of Zeeman splitting, back-projection effects (see following paragraph) and Dingle damping, which strongly damps higher frequencies at low fields.

Since the magnetisation increases linearly up to \( H_c \)[3], we would expect only smooth changes in the quantum oscillation spectrum as a function of magnetic field when the Fermi surface is spin-split. However, a smooth change in the Fermi surface can appear as an abrupt change in the quantum oscillation frequency due to the back projection effect \[12\], an effective Doppler shift of the observed frequency, \( F_{\text{obs}} \), with respect to the true frequency, \( F_{\text{true}} \), such that \( F_{\text{obs}} = F_{\text{true}}(B) - B \frac{dF_{\text{true}}(B)}{dB} \). Hence, if \( F_{\text{true}} \) changes only linearly with \( B \) up to \( H_c \), no change would be seen in \( F_{\text{obs}} \) until \( H_c \) is crossed.

4. Conclusion
We have measured the magnetostriction in GdSb up to 55 T with a new high resolution technique. The improved resolution allows us to resolve quantum oscillations in the magnetostriction both above and below the critical field. We observe a change in the quantum oscillation spectrum coincident with the switching of the sense of the magnetoelastic coupling to the local moment system. This may either be a consequence of magnetic reconstruction of the Fermi surface, or of Zeeman splitting largely hidden by the subtleties of quantum oscillation measurements. It is difficult, given the signal-to-noise ratio and small number of oscillations involved, to differentiate between these two scenarios; higher resolution quantum oscillation measurements are required.
**Figure 2.** a) The oscillatory magnetostriction of GdSb at 2.1 K as a function of inverse field. The saturation field $H_s$ is indicated by the vertical grey line. b) Fourier transform of the data for the fixed field range 34–54 T for several temperatures using a Hann window to eliminate sidebands. Several frequencies $F_{1-4}$ are present. $F_1^-$ is likely to be an artifact as its amplitude is highly dependent on the order of polynomial background subtraction that is used. c) Fourier transforms for the field range between 15 T and $H_c$. Only one frequency is clearly present, $F_0 = 360 \pm 20$ T. The change in frequency spectrum on crossing $H_c$ is discussed in the text.

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