Modeling magnetostriction in RCu$_2$ compounds using \textit{McPhase}

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In RCu$_2$ compounds (R = rare earth) magnetostriction results from two different sources, the single ion crystal field contribution and the two ion exchange interaction. The crystal field contribution can also be observed above the magnetic ordering temperature and leads to giant magnetostriction in high magnetic fields at some members of the series. The exchange contribution has to be considered additionally when describing the magnetostriction in the magnetically ordered state. A new feature of the mean field–Monte Carlo simulation program \textit{McPhase} (http://www.mcphase.de) allows to calculate the magnetic phase diagram and model the exchange contribution. We present new data of thermal expansion and the longitudinal and transversal magnetostriction of NdCu$_2$ with magnetic field applied along the $a$, $b$, and $c$ direction of the orthorhombic single crystal. These data are compared to the results of a \textit{McPhase} simulation, which is based on exchange parameters derived from measurements of the magnetic excitations. The magnetoelastic interaction is compared to the case of GdCu$_2$. The analysis can be extended to other RCu$_2$ compounds. © 2002 American Institute of Physics. [DOI: 10.1063/1.1449363]

I. MAGNETOElastic INTERACTIONS IN RCu$_2$ COMPOUNDS

According to Ref. 1 the static and dynamic magnetic properties of RCu$_2$ compounds in the magnetically ordered state can be described by a Hamiltonian, which is the sum of crystal field (CF), exchange (EX), and Zeeman contributions and the elastic energy

$$\mathcal{H} = H_{\text{CF}} + H_{\text{EX}} - \sum_i \frac{g \mu_B H}{2} J_i + E_{\text{el}}. \quad (1.1)$$

In order to describe magnetoelastic properties the strain dependence of the CF for ear in the strain tensor $\epsilon_{aa}$ can be derived. This is done for the special case of orthorhombic RCu$_2$ compounds with ferromagnetic $bc$ planes (such as NdCu$_2$) and considering the strain dependence of the CF for $l=2$ and of the EX up to the next nearest $bc$ plane. The following expressions are calculated using a mean field model, expanding the CF and EX parameters linear in the strain tensor $\epsilon$ and minimizing the free energy with respect to $\epsilon$ similar to \textsuperscript{3,5}

$$\epsilon_{aa} = \epsilon_{\text{CF}}^{aa} + \epsilon_{\text{EX}}^{aa}. \quad (1.4)$$

\textbf{TABLE I.} Magnetoelastic parameters used in the evaluation of the strains by Eq. (1.5).

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>$a$</th>
<th>$b$</th>
<th>$c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_{aa}$</td>
<td>$-5 \times 10^{-5}$</td>
<td>$7 \times 10^{-5}$</td>
<td>$-8 \times 10^{-6}$</td>
</tr>
<tr>
<td>$B_{aa}$</td>
<td>$-5 \times 10^{-5}$</td>
<td>$3 \times 10^{-4}$</td>
<td>$2 \times 10^{-4}$</td>
</tr>
<tr>
<td>$K_{aa}$</td>
<td>$+4 \times 10^{-4}$</td>
<td>$-1 \times 10^{-4}$</td>
<td>$+2 \times 10^{-5}$</td>
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<tr>
<td>$K_{ab}$</td>
<td>$+5 \times 10^{-6}$</td>
<td>$-6 \times 10^{-5}$</td>
<td>$-4 \times 10^{-5}$</td>
</tr>
<tr>
<td>$K_{ac}$</td>
<td>$-1 \times 10^{-4}$</td>
<td>$-4 \times 10^{-4}$</td>
<td>$-1.8 \times 10^{-4}$</td>
</tr>
<tr>
<td>$L_{aa}$</td>
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<td>$+2 \times 10^{-4}$</td>
<td>$-1.3 \times 10^{-5}$</td>
</tr>
<tr>
<td>$L_{ab}$</td>
<td>$-15 \times 10^{-5}$</td>
<td>$-4 \times 10^{-4}$</td>
<td>$-1.8 \times 10^{-4}$</td>
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<td>$M_{aa}$</td>
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<td>$-1.9 \times 10^{-5}$</td>
<td></td>
</tr>
<tr>
<td>$M_{ab}$</td>
<td>$+6 \times 10^{-6}$</td>
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</table>

\textsuperscript{a}Values taken from Ref. 5.
magnetic excitations. The CF contribution \( \epsilon^{CF}_{aa} \) in Eq. (1.5) was calculated using the mean fields calculated with McPhase and the CF parameters given in Refs. 1 and 5.

II. RESULTS AND DISCUSSION

At low temperatures the contribution of phonons and electrons to the strain is negligible and the magnetostriction of NdCu2 can be calculated using the model (1.5). Table I shows the magnetoelastic parameters used in our evaluation of Eq. (1.5). The values for \( A \) and \( B \) have been taken from Ref. 5. The magnetoelastic constants \( K, L, \) and \( M \) have been determined to give a reasonable quantitative adaptation of the calculated expressions (1.5) to the experimental data. Interactions beyond the nearest neighbor (i.e., \( L,M \neq 0 \)) have only been taken into account when necessary in order to describe the experimental data. Magnetostriiction data on GdCu2 (see Ref. 2) yield \( K^{GdCu2}_{aa} = +17 \times 10^{-5} \), \( K^{GdCu2}_{ab} = -1 \times 10^{-5} \), and \( K^{GdCu2}_{ac} = -1 \times 10^{-5} \) (\( a = a,b,c \)). Comparing these values to NdCu2 we infer that in the heavy RCu2 compounds the exchange contribution to the magnetostriction is of comparable magnitude. Note that in the case of GdCu2 \( K_{aa} = K_{ab} = K_{ac} \) \( (a = a,b,c) \) in contrast to NdCu2. Probably the orbital momentum (\( L \neq 0 \)) of the Nd\(^{3+} \) ion leads to the strong anisotropy in the derivatives of the exchange tensor \( J_{\alpha \beta} \) with respect to the strain \( \epsilon \).

The results of the calculation are compared in Figs. 1 and 2 to new experimental data measured by capacitance dilatometry on a single crystal. The magnetostriiction data are in good agreement with a previous study of \( \Delta b/b \) for magnetic field in the \( b \) direction. The solid lines correspond to results of the McPhase calculation taking into account both, the exchange (\( \epsilon^{EX}_{aa} \)) and CF (\( \epsilon^{CF}_{aa} \)) contribution to the strain.

For comparison the CF contribution \( \epsilon^{CF}_{aa} \) is shown separately (long dashes). Although magnetic order has been taken into account, this contribution alone cannot describe our experimental data adequately. In order to show the influence of magnetic order, \( \epsilon^{CF}_{aa} \) has also been calculated assuming that the system stays paramagnetic down to zero temperature [i.e., by putting all \( J_{\alpha \beta i} = 0 \) in Eq. (1.3) and calculating the \( \langle O^{m}_{i}(\mathbf{J}) \rangle_{T,H} \) in Eq. (1.5) using the CF eigenstates].
The results of this calculation are shown by short dashes in Figs. 1 and 2.

The onset of the axis conversion in the c direction difficult (Fig. 1) and therefore values given for $K_{ac}$ are less reliable than the other magnetoelastic constants given in Table I.

In conclusion, the present analysis shows that it is possible to model magnetostriction in compounds with very complex magnetic phase diagrams using McPhase. Good agreement between experiment and theory can be achieved, if the parameters in the Hamiltonian are well determined. Further improvements of ab initio calculations may eventually lead to estimates of the magnetoelastic parameters.

In the case of NdCu$_2$ the CF contribution calculated according to Ref. 5 can only describe part of the experimental data. In the magnetically ordered phase it is important to consider the exchange contribution to the strain, which can be calculated by McPhase. The remaining discrepancies (e.g., the slope of $\Delta b/b$ and $\Delta c/c$ in high fields parallel a) suggest that possibly magnetoelastic CF contributions for $l>2$ are important. Furthermore, the CF model for small strains is not sufficient to describe the giant magnetostriction for high fields parallel c. This is probably due to the importance of large quadrupolar interactions which leads to a magnetic axis conversion.

In contrast to the rare earth elements in the R Cu$_2$ series the exchange contribution to the magnetostriction is of the same order as the single ion contribution. In order to establish more details of the exchange contribution it is necessary to extend the analysis of NdCu$_2$ and GdCu$_2$ to other R Cu$_2$ compounds.

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