Modeling magnetostriction in RCu_2 compounds using *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 *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 *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

$$\mathcal{H}=\mathcal{H}_{\mathrm{CF}}+\mathcal{H}_{\mathrm{EX}}-\sum_{\mathbf{i}} g_J\mu_B\mathbf{H}\cdot\mathbf{J_i}+E_{\mathrm{el}}. \tag{1.1}$$

In order to describe magnetoelastic properties the strain dependence of the CF and EX part has to be considered (for the notation see Ref. 3):

$$\mathcal{H}_{\mathrm{CF}}=\sum_{lm,\mathbf{i}} B_l^m(\boldsymbol{\epsilon})O_l^m(\mathbf{J_i}), \tag{1.2}$$

$$\mathcal{H}_{\mathrm{EX}}=-\frac{1}{2}\sum_{\mathbf{ij},\alpha\beta}\mathbf{J}_\mathbf{i}^\alpha\mathcal{J}_{\alpha\beta}(\boldsymbol{\epsilon},\mathbf{i}-\mathbf{j})\mathbf{J}_\mathbf{j}^\beta, \tag{1.3}$$

$$E_{\mathrm{el}}=\frac{1}{2}\sum_{\alpha\beta\alpha'\beta'}\boldsymbol{\epsilon}_{\alpha\beta}\boldsymbol{\epsilon}_{\alpha'\beta'}c^{\alpha\beta\alpha'\beta'}. \tag{1.4}$$

From this model the diagonal components of the strain tensor $\boldsymbol{\epsilon}$ 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 $\boldsymbol{\epsilon}$ and minimizing the free energy with respect to $\boldsymbol{\epsilon}$ similar to[4,5]

$$\boldsymbol{\epsilon}_{\alpha\alpha}=\boldsymbol{\epsilon}_{\alpha\alpha}^{\mathrm{CF}}+\boldsymbol{\epsilon}_{\alpha\alpha}^{\mathrm{EX}},$$

$$\boldsymbol{\epsilon}_{\alpha\alpha}^{\mathrm{CF}}=\frac{1}{N}\sum_{\mathbf{i}}\left[A_\alpha\langle O_2^0(\mathbf{J_i})\rangle_{T,\mathbf{H}}+B_\alpha\langle O_2^2(\mathbf{J_i})\rangle_{T,\mathbf{H}}\right], \tag{1.5}$$

$$\boldsymbol{\epsilon}_{\alpha\alpha}^{\mathrm{EX}}=\frac{1}{N}\sum_{\beta\mathbf{i}}\left[K_{\alpha\beta}\langle\mathbf{J}_\mathbf{i}^\beta\mathbf{J}_\mathbf{i}^\beta\rangle_{T,\mathbf{H}}+L_{\alpha\beta}\langle\mathbf{J}_\mathbf{i}^\beta\mathbf{J}_{\mathbf{i}+\mathbf{a}/2+\mathbf{b}/2+\mathbf{c}/2}^\beta\rangle_{T,\mathbf{H}}+M_{\alpha\beta}\langle\mathbf{J}_\mathbf{i}^\beta\mathbf{J}_{\mathbf{i}+\mathbf{a}}^\beta\rangle_{T,\mathbf{H}}\right]. \tag{1.6}$$

$\langle\ \rangle_{T,\mathbf{H}}$ denote thermal expectation values, $\mathbf{i}$ the position vectors of the rare earth, $\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}$ the orthorhombic lattice vectors.

Using a new version of the *McPhase* simulation program it is possible to calculate the static correlation functions $\langle\mathbf{J}_\mathbf{i}^\beta\mathbf{J}_{\mathbf{i}'}^\beta\rangle_{T,\mathbf{H}}$ in Eq. (1.6). The complete $\mathbf{H}$ and T dependence of the strain is determined by the expectation values $\langle\ \rangle_{T,\mathbf{H}}$. The model used for the simulation of the magnetically ordered phases has been described in Ref. 6 for external fields parallel to b. Here we extend it to fields in a and c using the magnetic exchange determined from the dispersion of the

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

$\alpha=$	a	b	c
A_α [a]	-5×10^{-5}	$+7\times10^{-5}$	-8×10^{-6}
B_α [a]	-5×10^{-5}	$+3\times10^{-4}$	$+2\times10^{-4}$
$K_{\alpha a}$	$+4\times10^{-4}$	-1×10^{-4}	$+2\times10^{-5}$
$K_{\alpha b}$	$+5\times10^{-6}$	-6×10^{-5}	-4×10^{-5}
$K_{\alpha c}$	-1×10^{-4}	-4×10^{-4}	-1.8×10^{-4}
$L_{\alpha a}$			
$L_{\alpha b}$	$+1.5\times10^{-5}$		-1.3×10^{-5}
$L_{\alpha c}$			
$M_{\alpha a}$			
$M_{\alpha b}$	$+6\times10^{-6}$	-1.9×10^{-5}	
$M_{\alpha c}$			

[a]Values taken from Ref. 5.

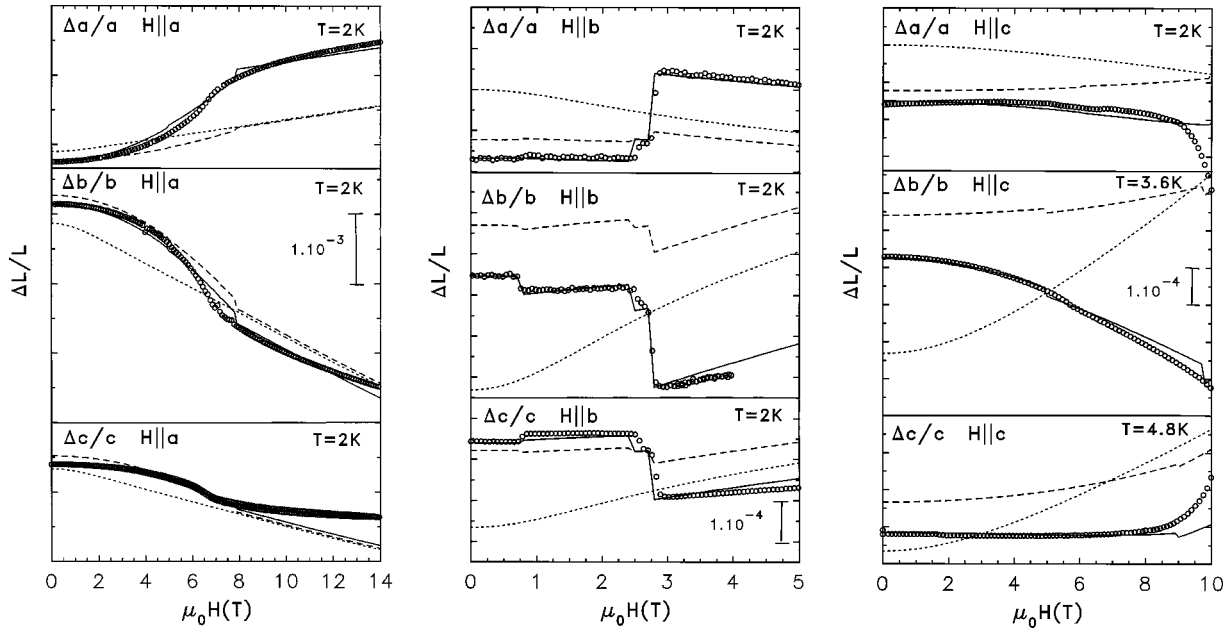

FIG. 1. Longitudinal and transversal magnetostriction of a $NdCu_2$ single crystal. The symbols indicate the measured data. The solid lines (—) correspond to results of the *McPhase* calculation taking into account both the exchange contribution $\epsilon_{\alpha\alpha}^{\mathrm{EX}}$ and the CF contribution $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ to the strain. For comparison the CF contribution $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ is shown separately (long dashes – – –). Short dashes (---) indicate $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ calculated without *McPhase*, i.e., considering only the paramagnetic CF eigenstates (see text).

magnetic excitations.[1] The CF contribution $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ 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 $NdCu_2$ 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. Magnetostriction data on $GdCu_2$ (see Ref. 2) yield $K_{a\alpha}^{\mathrm{GdCu_2}}=+17\times10^{-5}$, $K_{b\alpha}^{\mathrm{GdCu_2}}=-1\times10^{-5}$, and $K_{c\alpha}^{\mathrm{GdCu_2}}=-1\times10^{-5}$ $(\alpha=a,b,c)$. Comparing these values to $NdCu_2$ we infer that in the heavy RCu_2 compounds the exchange contribution to the magnetostriction is of comparable magnitude. Note that in the case of $GdCu_2$ $K_{\alpha a}=K_{\alpha b}=K_{\alpha c}$ $(\alpha=a,b,c)$ in contrast to $NdCu_2$. Probably the orbital momentum $(L\neq 0)$ of the Nd^{3+} ion leads to the strong anisotropy in the derivatives of the exchange tensor $\mathcal{J}_{\alpha\beta}$ with respect to the strain ϵ.

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 magnetostriction data are in good agreement with a previous study of $\Delta b/b$ for magnetic field in the b direction.[7] The solid lines correspond to results of the *McPhase* calculation taking into account both, the exchange $(\epsilon_{\alpha\alpha}^{\mathrm{EX}})$ and CF $(\epsilon_{\alpha\alpha}^{\mathrm{CF}})$ contribution to the strain. For comparison the CF contribution $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ 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, $\epsilon_{\alpha\alpha}^{\mathrm{CF}}$ has also been calculated assuming that the system stays paramagnetic down to zero temperature [i.e., by putting all $\mathcal{J}_{\alpha\beta}(\epsilon,\mathbf{i}-\mathbf{j})=0$ in Eq. (1.3) and calculating the $\langle O_l^m(\mathbf{J_i})\rangle_{T,\mathbf{H}}$ in Eq. (1.5) using the CF eigenstates].

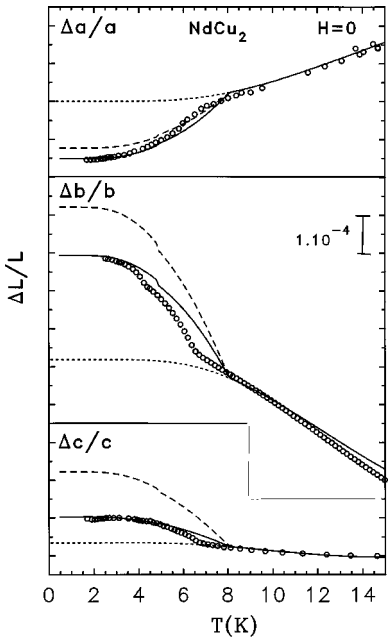

FIG. 2. Thermal expansion of a $NdCu_2$ single crystal in zero field in the crystallographic a, b, and c direction. For explanations of symbols see Fig. 1 and text.

The results of this calculation are shown by short dashes in Figs. 1 and 2.

The onset of the axis conversion[8] at around 8 T makes the interpretation of magnetostriction for fields in the c direction difficult (Fig. 1) and therefore values given for $K_{\alpha c}$ 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[9] 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[4] 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.[8]

In contrast to the rare earth elements[9] in the RCu_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 RCu_2 compounds.

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