MÖSSBAUER INVESTIGATION OF DEOXYMYOGLOBIN SINGLE CRYSTALS WITH POLARIZED γ-RAYS

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Deoxymyoglobin (MB) single crystals have been investigated using polarized (and unpolarized) $\gamma$-rays. The experimental values obtained with unpolarized radiation were in agreement with our former work\(^1\). As a polarized source we used Co\(^{57}\) (100 mCi) embedded in $\alpha$-iron at room temperature magnetized perpendicular to the $\gamma$-ray propagation direction. This arrangement yields six linear polarized lines, A, B, ..., F. Spectra have been recorded for different directions of the magnetization $\vec{M}$ relative to the crystal axis b and for different angles of rotation $\varphi$ around the b axis (Fig. 2). Some of the spectra are shown in Fig.1. The absorber transitions $\alpha$ and $\beta$, resulting from quadrupole interaction, combined with the six emission lines lead to twelve absorption lines. From the computer fits we got intensity ratios $A\beta/A\alpha$, $B\beta/B\alpha$ as a function of the angle $\gamma$.

To determine the EFG parameters from the measured intensity ratios a method on the basis of the density matrix formalism has been worked out. We explicitly calculated the density matrix $\hat{\rho}_\theta$ for transitions in Fe\(^{57}\) nuclei. Using the formula\(^2\) for the resonantly absorbed intensity $I = \text{tr}(\hat{\rho}_\theta \cdot \hat{\rho}_\Theta)$ (in the thin absorber approximation) where $\hat{\rho}_\Theta$ is the density matrix of the source radiation, we derive theoretical intensity ratios for different polarizations of the source radiation as a function of the rotation angle.

Applied to our experimental data the formulæ lead to a system of equations for angles $\alpha_\Theta$, $\beta_\Theta$, $\Theta$, which depend on the EFG orientation relative to the axes system $(x,y,z)$ (s. Fig.2), the asymmetry parameter $\eta$ and the sign of the main component $V_{zz}$ of the EFG. Thus only a manifold of solutions can be obtained (Fig.3). This ambiguity is due to the fact that in MB single crystals we have two iron atoms per unit cell, one atom being transformed into the other by a 180°-rotation around the b axis. In the case of only one atom per unit cell, with polarized radiation the EFG parameters could be uniquely determined by rotation around one axis whereas with an unpolarized source one would need (generally) rotations around two non-collinear axes.

As a consequence of the theoretical treatment the intensity ratios for unpolarized $\gamma$-rays and for x-polarized $\gamma$-rays are strictly correlated and the x-polarized ratios are constant (with value $k$) for rotations around the y axis, which is in our case parallel to the b axis (s. Fig.2). This is well represented by our experiments, but the measured value $K=1.00\pm0.1$ is different from the theoretically expected value, which should be 0.65 to be consistent with the intensity ratios obtained with unpolarized radiation. The discrepancy of the theoretical and experimental $k$ values is only slightly improved by taking into account a deficiency of source polarization, thickness effects, angle uncertainties etc. However, an explanation might be given by assuming that a pronounced anisotropic relaxation effect is operative similar to observations made by Morup\(^3\).

References:
Fig. 1: $\vec{M}$ is the direction of the source magnetization, $b$ is the twofold symmetry axis of the single crystal. The angles given in the picture specify the rotation around the $b$ axis. For all spectra, $\vec{M}$ and $b$ were perpendicular to the $\gamma$-ray, and $T$ was 77 K. Note that the line positions are all known from appropriate combination of emission lines $A, ..., F$ with the absorber transitions $\alpha$ and $\beta$.

Fig. 2: Schematic of the orientation relations in the experiments. $\gamma=250^\circ$ corresponds to $\gamma$-ray $/c$, and $\gamma=340^\circ$ to $\gamma$-ray $/a'$. Supported by Deutsche Forschungsgemeinschaft.

Fig. 3: Manyfold of solutions obtained by our theoretical analysis of the experimental data. The angles $\alpha_0$, $\beta_0$, $\theta_{\min(\max)}$ describe the transformation of the laboratory system to the EFG system in the case where the experimentally obtained intensity ratio $R=I_b/I_\alpha$ for unpolarized $\gamma$-rays is minimal (maximal). The solid lines (unprimed angles) correspond to $R_{\min}=0.66$, $R_{\max}=1.18$ and $k=0.65$, where $k$ is the constant ratio for $x$-polarized $\gamma$-rays. The measured values were $R_{\min}=0.67\pm0.07$, $R_{\max}=1.18\pm0.11$ and $k=1.0\pm0.11$. If we keep $k=1.0$ the dashed lines (primed angles) are obtained for $R_{\min}=0.76$ and $R_{\max}=1.31$. 

$\theta_0$ and $\theta_0'$ are the angles between the $\gamma$-ray and the $b$ axis in the laboratory and EFG systems, respectively. The measured values were $\theta_0=40^\circ$ and $\theta_0'=50^\circ$. The measured values were $\theta_{\min}=30^\circ$ and $\theta_{\max}=60^\circ$. If we keep $\theta_0=40^\circ$ the dashed lines (primed angles) are obtained for $\theta_{\min}=35^\circ$ and $\theta_{\max}=55^\circ$. Supported by Deutsche Forschungsgemeinschaft.