IS IT POSSIBLE TO STUDY NANOCRYSTAL FERROMAGNETIC FILMS BY µSR-TECHNIQUE?

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Main features of the muon spin polarization behavior in nanostructured films

A behaviour of a muon spin polarization strongly depend on a behaviour of a muon itself. Note first of all that a muon can stop both in some interstitial site of a grain and in an intergrain area. So, we can write a muon spin polarization in a form of a sum

$$\mathcal{P}(t) = \mathbf{P}_{\rm cr}(t) + \mathbf{P}_{\rm nc}(t), \qquad (28)$$

where $\mathbf{P}_{cr}(t)$ and $\mathbf{P}_{nc}(t)$ are fraction of a polarization in a grain and outside of it respectively.

For non-diffusion muons in disordered media we have the well-known formula

$$P_i(t) = \mu_{ik}(t)P_k(0)$$

where the polarization tensor is determined as

$$\mu_{ik}(t) = n_i n_k + (\delta_{ik} - n_i n_k) \cos \gamma b_\mu t + e_{ikl} n_l \sin \gamma b_\mu t$$

Here

 \mathbf{b}_{μ} is the local field at the muon position $\mathbf{n} = \mathbf{b}_{\mu}/|\mathbf{b}_{\mu}|$ - the unit vector and $\gamma = 13.554 \text{ kHz/G}$ - gyromagnetic ratio for the muon

Hierarchy of Fields

 \mathcal{B}_{\cdot} - an external field to the whole sample (film); macroscopic fields \mathbf{B} and \mathbf{H} and a magnetization M inside grains. Averaged fields $\langle \mathbf{B} \rangle$, $\langle \mathbf{H} \rangle$ and a magnetization $\langle \mathbf{M} \rangle$. in a film. $\mathcal{B}_i = \langle H_i \rangle + 4\pi N_{ik} \langle M_k \rangle, \quad \langle \mathbf{B} \rangle = \langle \mathbf{H} \rangle + 4\pi \langle \mathbf{M} \rangle.$ (1) $\langle \mathbf{B} \rangle, \langle \mathbf{H} \rangle$ and $\langle \mathbf{M} \rangle$ fields and magnetization in an intergrain volume; a field $\langle \mathbf{B} \rangle$ acts on a muon stopped outside a grain. $N_{zz} \equiv N_{\perp} \approx 1, \quad N_{\parallel} \sim N_{xx} \sim N_{yy} \sim N_{xy} \ll 1.$ $N_{zx} \sim N_{zy} \ll 1.$ $\langle \mathbf{B} \rangle$ is the "external" field for grains.



Рис. 1:

External field is directed perpendicular to the plane

$$\mathcal{B}_z \equiv \mathcal{B} = \langle H_z \rangle + 4\pi \langle M_z \rangle, \quad \langle H_{\rm pl} \rangle \approx 0,$$

 $\mathcal{B} \| z$

$$\langle B_z \rangle = \mathcal{B}, \quad \langle B_{\rm pl} \rangle \approx 4\pi \langle M_{\rm pl} \rangle.$$

External field is parallel to the plane $\mathcal{B} \perp z$,

$$\langle H_z \rangle + 4\pi \langle M_z \rangle = 0, \quad \langle H_z \rangle = -4\pi \langle M_z \rangle, \quad \langle H_{\rm pl} \rangle \approx \mathcal{B}.$$

 $\langle B_z \rangle = 4\pi \langle M_z \rangle, \quad \langle B_{\rm pl} \rangle = \mathcal{B} + 4\pi \langle M_{\rm pl} \rangle.$

For the every grain $\langle B_i \rangle = H_i + 4\pi n_{ik} M_k$,

$$n_{\parallel} = n_{zz} \approx 1, \quad n_{\perp} = 1 - n_{\parallel} \ll 1,$$

External field is directed perpendicular to the plane

$$\langle B_z \rangle \approx \mathcal{B} = H_z + 4\pi M_z,$$
 или $H_z = \mathcal{B} - 4\pi M_z.$
 $H_{\rm pl} = \langle B_{\rm pl} \rangle = 4\pi \langle M_{\rm pl} \rangle, \quad B_{\rm pl} = 4\pi (\langle M_{\rm pl} \rangle + M_{\rm pl}).$

 $\mathcal{B}||z|$

External field is parallel to the plane
$$\mathcal{B} \perp z$$
.

$$H_z = -4\pi M_z, \quad H_{\rm pl} \approx \langle B_{\rm pl} \rangle = \mathcal{B} + 4\pi \langle M_{\rm pl} \rangle.$$
$$B_z = 0, \quad B_{\rm pl} = \mathcal{B} + 4\pi (\langle M_{\rm pl} \rangle + M_{\rm pl}).$$

We need to determine **M** inside grains and *<***M***>* inside the film.

For a single-axis ferromagnet

$$U_{\rm an} = K_{ik} m_i m_k,$$

Minimization of free-energy potential for a grain

$$\widetilde{F} = \frac{1}{2} (4\pi n_{ik} + \beta_{ik}) M_i M_k - \langle \mathbf{B} \rangle \mathbf{M},$$

For $\mathcal{B} || z$
$$\widetilde{F}(\theta) = \frac{\beta}{2} M^2 \sin^2 \theta + 2\pi M^2 \cos^2(\vartheta - \theta) - M\mathcal{B} \cos(\vartheta - \theta)$$

Minimum condition

$$\frac{1}{2}\beta M\sin 2\theta - 2\pi M\sin 2(\vartheta - \theta) + \mathcal{B}\sin(\vartheta - \theta) = 0.$$

Analytical solution in a limit of strong external fields

$$\mathcal{B} \gg M$$
, or $|\langle \mathbf{B} \rangle| \gg M$

Define $\theta = \vartheta - \delta_{1}$

$$\delta \approx \frac{1}{2} \beta \frac{M}{\mathcal{B}} \sin 2\vartheta.$$

Inside grains

$$\begin{split} M_z &= M\cos\delta \approx M \left(1 - \frac{\beta^2}{8} \frac{M^2}{\mathcal{B}^2} \sin^2 2\vartheta \right) \\ M_{\rm pl} &= M\sin\delta \approx \frac{1}{2} \beta \frac{M^2}{\mathcal{B}} \sin 2\vartheta. \end{split}$$

Respectively

$$\langle M_z \rangle = M \left(1 - \frac{\beta^2}{15} \frac{M^2}{\mathcal{B}^2} \right), \quad \langle M_{\rm pl} \rangle = 0.$$

For $\mathcal{B} \perp z$, we have:

$$M_z = M \sin \delta, \quad \langle M_z \rangle = 0, \quad M_{\rm pl} = M \cos \delta, \quad \langle M_{\rm pl} \rangle = M \left(1 - \frac{\beta^2}{15} \frac{M^2}{\mathcal{B}^2} \right).$$

For cubic ferromagnet of easy-axis type

$$\widetilde{F} = -\frac{1}{4}\beta M^2 \left(m_{\xi}^4 + m_{\eta}^4 + m_{\zeta}^4 \right) + 2\pi n_{ik} M_i M_k - \langle \mathbf{B} \rangle \mathbf{M}.$$

Direction of the magnetization vector

$$\delta \approx \frac{1}{4} \beta \frac{M}{\mathcal{B}} \sin 4\vartheta.$$

For the external field $\mathcal{B}||z|$

$$\begin{split} M_z &= M\cos\delta \approx M\left(1 - \frac{\beta^2}{32}\frac{M^2}{\mathcal{B}^2}\sin^2 4\vartheta\right),\\ M_{\rm pl} &= M\sin\delta \approx \frac{1}{4}\beta\frac{M^2}{\mathcal{B}}\sin 4\vartheta.\\ \langle M_z \rangle &= M\left(1 - \frac{\beta^2}{63}\frac{M^2}{\mathcal{B}^2}\right), \quad \langle M_{\rm pl} \rangle = 0. \end{split}$$

If the external field $\mathcal{B} \perp z$,

 $M_z = -M\sin\delta, \quad \langle M_z \rangle = 0, \quad M_{\rm pl} = M\cos\delta, \quad \langle M_{\rm pl} \rangle = M\left(1 - \frac{\beta^2}{63}\frac{M^2}{\mathcal{B}^2}\right).$

Local field at a muon

Local field at a muon determines a precession frequency of its spin polarization. A local field at a muon in any grain is determined by the formula [14, 15]

$$\mathbf{b}_{\mu} = \mathbf{B} - \frac{8\pi}{3}\mathbf{M} + \mathbf{b}_{dip} + \mathbf{B}_{cont}, \qquad (18)$$

where \mathbf{B}_{cont} is the field created by conduction electrons, and \mathbf{b}_{dip} is the microscopic field of all dipoles inside the Lorentz sphere. A contact field could always written in the form

$$B_{i\,\mathrm{cont}} = K_{ik}B_k.\tag{19}$$

For cubic crystals is valid a relation $K_{ik} = \delta_{ik}K$ and contact field is reduced to the well-known isotropic Knight shift. Microscopic field \mathbf{b}_{dip} inside the Lorentz sphere one can represent in a form [14, 15])

$$b_{i\,\rm dip} = -\frac{4\pi}{3}M_i + a_{ik}M_k.$$
 (20)

A tensor a_{ik} depends on an interstitial site where a muon stopped. It can be calculated by the well-known Evald method. Calculations shown (see e.g. [14, 15]) that for the FCC-lattice (Ni) the tensor is determined as $a_{ik} = \delta_{ik} 4\pi/3$, hence, a microscopic dipole field is equal to zero:

$$\mathbf{b}_{\rm dip}(fcc) = 0. \tag{21}$$

Thus, for the FCC-lattice the local field at a muon doesn't depend on a type of an interstitial site and is equal to

$$\mathbf{b}_{\mu}(fcc) = \mathbf{B} - \frac{8\pi}{3}\mathbf{M}.$$
(22)

Microscopic dipole field for the HCP-lattice (Co) is small too, but has different values for different crystallographically unequivalent interstitial sites:

$$a_{ik}(hcp) = \frac{4\pi}{3} + \delta a_{ik}.$$
(23)

If the z-axes is directed along the hexagonal axes components of the tensor a_{ik} can be written in the form:

$$\begin{aligned} \delta a_{xx}^h &= \delta a_{yy}^h = \delta/2, \quad \delta a_{zz}^h = -\delta & \text{in an octahedral site,} \\ \delta a_{xx}^t &= \delta a_{yy}^t = -\delta, \quad \delta a_{zz}^t = 2\delta & \text{in a tetrahedral site.} \end{aligned} \tag{24}$$

We can consider $b_{dip} \ll M$ because $\delta \approx 0.104$. The local field at a muon in the HCP-lattice can be written in the form:

$$b_{\mu i}(hcp) = B_i - \frac{8\pi}{3}M_i + \delta a_{ik}M_k.$$
 (25)

The more complicated picture is observed in the BCC-lattice (Fe) where a dipole field have a large value and depend both on a type of an interstitial site and on a direction of a magnetization vector **M**. Components of a tensor $a_{ik}(bcc)$ in the main axis are equal to

$$\begin{array}{ll}
a_{xx}^h &= a_{yy}^h = -1.165, & a_{zz}^h = 14.9 & \text{in an octahedral site,} \\
a_{xx}^t &= a_{yy}^t = 5.707, & a_{zz}^t = 1.152 & \text{in a tetrahedral site.} \\
\end{array} \tag{26}$$

The local field at a muon in the BCC-lattice can be written in the form:

$$b_{\mu i}(bcc) = B_i - 4\pi M_i + a_{ik}(bcc)M_k.$$
 (27)

Fast diffusion

No difference between HCP and BCC lattices. Strong fields, $\mathcal{B}||z|$ Local field:

$$\begin{split} b_z = &(1+K)\mathcal{B} - \frac{8\pi}{3}M_z \approx \mathcal{B} - \frac{8\pi}{3}M\left(1 - \frac{1}{2}\delta^2\right),\\ b_{\rm pl} = &(1+K)B_{\rm pl} - \frac{8\pi}{3}M_{\rm pl} \approx \frac{4\pi}{3}M\delta, \end{split}$$

Two items for the local field:

$$b = b_0 + b(\vartheta).$$

$$b_0 = \mathcal{B} - \frac{8\pi}{3}M + \frac{1}{2}\left(\frac{4\pi}{3}\right)^2 \frac{M^2}{\mathcal{B}}, \quad b(\vartheta) = \frac{4\pi}{3}M\delta^2.$$

Transverse polarization

$$\mathcal{P}_{\perp} = \langle \frac{b_z}{b} e^{-i\gamma_{\mu}bt} \rangle \approx e^{-i\omega_0 t} \langle e^{-i\omega(\vartheta)t} \rangle,$$
$$\omega_0 = \gamma_{\mu} b_0, \ \omega(\vartheta) = \gamma_{\mu} b(\vartheta).$$

$$\mathcal{P}_{\perp}^{\text{diff}} = \mathrm{e}^{-\mathrm{i}(\omega_0 + \Delta\omega)t} \mathrm{e}^{-\sigma_{\text{diff}}^2 t^2},$$

$$\Delta \omega = \gamma_{\mu} \frac{1}{63} \frac{2\pi}{3} M \left(\frac{M}{\mathcal{B}}\right)^2 \quad \sigma_{\text{diff}}^2 = \frac{1}{2} \left(\frac{4\pi}{3} \gamma_{\mu} M\right)^2 \langle \delta^2 \rangle$$

Longitudinal polarization

$$\mathcal{P}_{\parallel}^{\text{diff}} = \mathrm{e}^{-\mathrm{i}\omega_{0}t} \langle \frac{b_{\mathrm{pl}}}{b} \mathrm{e}^{-\mathrm{i}\omega(\vartheta)t} \rangle = 0.$$

Non-diffusion muons

BCC lattice, $\mathcal{B} \| z$

Components of the dipolar tensor

$$a_{zz} = a + \delta a \cos 2\vartheta, \quad a_{xx} = a - \delta a \cos 2\vartheta, \quad a_{zx} = a_{xz} = \delta a \sin 2\vartheta,$$

where

$$a = (a_{\parallel} + a_{\perp})/2, \ \delta a = (a_{\parallel} - a_{\perp})/2, \ a_{\parallel} = a_{\zeta\zeta}, \ a_{\perp} = a_{\xi\xi}.$$

Components of the local field:

$$\begin{split} b_{z} &= \mathcal{B} - 4\pi M_{z} + a_{zz} M_{z} + a_{zx} M_{\text{pl}} = \\ &= \mathcal{B} - \frac{1}{2} (4\pi + a_{\perp}) M + \delta a M \cos 2\vartheta + \frac{1}{4} \delta a \beta \frac{M^{2}}{\mathcal{B}} \sin 4\vartheta \cos 2\vartheta, \\ b_{\text{pl}} &= \delta a M \sin 2\vartheta - \left[\frac{1}{2} (4\pi + a_{\perp}) + \delta a \cos 2\vartheta \right] \frac{1}{4} \beta \frac{M^{2}}{\mathcal{B}} \sin 4\vartheta. \end{split}$$

Two items of the local field:

$$b_{0} = \mathcal{B} - \frac{1}{2}(4\pi + a_{\perp})M,$$

$$b(\vartheta) \approx \delta a M \cos 2\vartheta + \frac{1}{2}\delta a \left(\frac{1}{4}\beta \cos 2\vartheta + \delta a\right) \frac{M^{2}}{\mathcal{B}} \sin^{2} 2\vartheta.$$

Transverse component of a polarization

$$\mathcal{P}_{\perp} = \langle \frac{b_z}{b} \mathrm{e}^{-\mathrm{i}\omega(\vartheta)t} \rangle \mathrm{e}^{-\mathrm{i}\omega_0 t}.$$

represented by the gaussian exponent

$$\mathcal{P}_{\perp} = \mathrm{e}^{-\mathrm{i}(\omega_0 + \Delta\omega)t} \mathrm{e}^{-\sigma_{\mathrm{nd}}^2 t^2},$$

$$\begin{split} \omega_0 &= \gamma_\mu \mathcal{B} \left(1 - (2\pi + \frac{1}{2}a_\perp)\frac{M}{\mathcal{B}} \right), \\ \Delta \omega &= -\frac{1}{3}\gamma_\mu \delta a M \left[1 - \frac{2}{15} \left(2\delta a - \frac{41}{28}\beta \right) \frac{M}{\mathcal{B}} \right], \\ \sigma_{\rm nd}^2 &= \frac{7}{30} \left(\gamma_\mu \delta a M \right)^2. \end{split}$$

Longitudinal polarization is equal approximately to zero

$$\mathcal{P}_{\parallel} = \langle \frac{b_{\mathrm{pl}}}{b} \mathrm{e}^{-\mathrm{i}\omega(\vartheta)t} \rangle \mathrm{e}^{-\mathrm{i}\omega_0 t} = 0.$$

For the external field parallel to the plane we have the same formulas.

In the case of single-axes ferromagnets we have a similar behavior:

- 1. No oscillations in the longitudinal polarization
- 2. Gaussian exponent for the transverse component of polarization with parameters

$$\begin{split} \omega_0^{hcp} &= \gamma_\mu \mathcal{B} \left[1 - \left(\frac{8\pi}{3} - \frac{1}{2} \delta a^{hcp} \right) \frac{M}{\mathcal{B}} \right], \\ \Delta \omega^{hcp} &= -\frac{1}{2} \gamma_\mu \delta a^{hcp} M \left[1 - \frac{4}{5} \left(\beta + \frac{3}{2} \delta a^{hcp} \right) \frac{M}{\mathcal{B}} \right], \\ \sigma_{\rm nd}^2 &= \frac{21}{40} \left(\gamma_\mu \delta a^{hcp} M \right)^2. \end{split}$$

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