Use of the \( \mu \)SR-technique for studying magnetization processes in nanocrystal films of ferromagnetic metals

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Abstract. I show that the \( \mu \)SR technique allows one to study the occurrence of a magnetic ordering in nanocrystal films made of ferromagnetic metals – so called “scale” phase transition. The relation between the microscopic (local) field with macroscopic characteristics just as the external magnetic field, the average magnetization and the saturation magnetization is determined for a model for which the nanocrystal film consists of crystallographically ordered grains separated by disordered areas. Expressions for the behaviour of the muon spin polarization ensemble in this type of structures are obtained in cases of fast diffusing and nondiffusing muons. It is shown that experiments with “slow” positively charged muons allow one to measure all parameters of this type of structures and obtain important information for the study of phase transition physics.

1. Introduction
Magnetic nanocrystalline metals are of interest for the \( \mu \)SR technique since the use of positive muons and neutron diffraction provides real possibilities to study their bulk properties\(^1\). Possible applications of the \( \mu \)SR technique for the study of nanocrystalline ferromagnetic materials were still mentioned few years ago \([1]\). Nevertheless, no serious experimental researches has been carried out yet. It was emphasized that at least two problems regarding the fundamental physics of magnets could be solved for nanostructured ferromagnets. One of them is the problem of specific “scale” phase transitions, when the existence of a spontaneous magnetization depends both on temperature and size of a crystal. The mechanism of this size-induced phase transition is being actively studied (see, e.g. \([2] - [4]\)). Currently, materials with grain sizes too small to exhibit ferromagnet properties are called superparamagnets. The second problem is related with the structure and magnetic properties of domains in nanostructures. They should be strongly different from the respective characteristics of “ordinary” polycrystalline ferromagnets. Actually, both theory and experiments show that microcrystalline grains \((10^3-4 \cdot 10^6 \text{ Å})\) should be single domain. The characteristic thickness of domain walls in a bulk sample\(^2\) is \(d \approx 10 - 30 \text{ Å}\), which is of the same order of magnitude as the thickness of the intercrystalline amorphous interface. Thus, the concept of magnetic domains and domain walls in nanocrystals differs from the one observed for macroscopic polycrystals. In our case we need to consider magnetized regions without domain grains in a nonmagnetic medium. Hence, the macroscopic field \(B_{\text{dom}}\)

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\(^1\) Nanocrystalline metals are conventionally accepted to be polycrystals with grain sizes of 10–400Å.

\(^2\) The characteristic thickness of domain walls depends on the ratio between exchange and magnetocrystalline anisotropy energy scale. The presented values are correct for the ferromagnets under consideration.
inside a crystallographically ordered grain and its dependence on the external magnetic field $B$ should differ significantly from the properties of macroscopic samples.

The possibilities to use the $\mu$SR technique for studying ferromagnetic nanostructured thin films are represented in this report.

2. Behaviour of muon spin polarization

Let consider a nanostructured film consisting of crystallographically ordered grains separated by disordered regions. Spontaneous magnetization can arise only in ordered regions. Therefore, the local field sensed by a muon depends on whether the muon stops in a site within a grain or in an intergrain region. Let consider the situation when a spontaneous magnetization does not equal to zero. Thus the spin polarization of the muons ensemble can be written as the sum

$$ P(t) = P_{cr}(t) + P_{nc}(t), $$

where $P_{cr}(t)$ and $P_{nc}(t)$ are the polarization of the muons fraction stopping in grains and in intergrain regions, respectively.

For non-diffusing muons, the polarization is given by the well-known expression (see e.g. [5])

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

where the tensor $\mu_{ik}(t)$ is

$$ \mu_{ik}(t) = n_in_k + (\delta_{ik} - n_in_k) \cos \gamma_{\mu}b_{\mu}t + e_{ikl}n_l \sin \gamma_{\mu}b_{\mu}t. $$

Here $b_{\mu}$ is the local field sensed by the muon, $\gamma_{\mu}/(2\pi) = 13.554$ kHz/G is the muon gyromagnetic ratio and $n = b_{\mu}/|b_{\mu}|$ is the unit vector along the magnetic field.

The local fields acting on a muon in a grain and an intergrain region differ significantly. We are first interested by the field in a crystallographically ordered grain. In general, by separating the Lorentz sphere around the muon position, we can write [5, 6]:

$$ b_{\mu} = B - \frac{8\pi}{3}M + b_{\text{dip}} + B_{\text{cont}}, $$

where $B_{\text{cont}}$ is the contact field induced by electrons and $b_{\text{dip}}$ is the microscopic field induced by the oriented magnetic dipoles inside the Lorentz sphere. The contact field can always be written as $B_{i\text{cont}} = K_{ik}B_k$. In cubic crystals, we can set $K_{ik} = \delta_{ik}K$. Therefore, the contact field causes only an isotropic Knight shift$^3$.

In an intergrain region, the spontaneous magnetization is equal to zero ($M_{\text{nc}} = 0$). Hence, dipole fields $b_{\text{dip}}$ can only be induced by the disordered nuclear magnetic moments. These fields cause an inhomogeneous line broadening, which can be adequately described by a Gaussian in the case of nondiffusing muons and a simple exponent for rapidly diffusing muons (see e.g. [5, 7]). Thus, the behaviour of the muon spin polarization for intergrain regions is controlled by the local magnetic field $b_{\mu} = \langle B \rangle + \delta b$, where $\delta b$ is the static field inhomogeneity. The characteristic scale of the field inhomogeneity in the intergrain region is determined by the magnetization of grains and the distance between them. Thus, the polarization precession frequency of muons in the intergrain fraction allows one to determine the average magnetic field in the film. In the case of nondiffusing muons a Gaussian damping allows to determine the characteristic scale of the field inhomogeneity, $\sigma \sim \gamma_{\mu}\langle \delta b^2 \rangle/\langle B \rangle$. In the case of diffusing muons, the depolarization rate depends on the diffusion constant and in the fast diffusing limit becomes negligibly small. It is

$^3$ For simplicity, we can omit the Knight shift in what follows, although it can make an appreciable contribution in some cases.
known that muons can diffuse rapidly in polycrystal samples (see e.g. [5] Ch.5 and references therein). But an irregular structure of an amorphous intergrain region differs from a structure of polycrystal samples. So, we can assume that a diffusion of a muon in intergrain regions is improbable.

If a crystallographically ordered grain has a nonzero spontaneous magnetization, the microscopic dipolar field is induced by the ordered electron magnetic moments. In this case, inside the Lorentz sphere, this field can always be written as [5, 6]

$$b_{\text{dip}} = -\frac{4\pi}{3} M_i + a_{ik}M_k,$$

where the tensor $a_{ik}$ depends on the type of interstitial sites. Calculations showed (see e.g. [5, 6]) that $a_{ik} = \delta_{ik}4\pi/3$ in an fcc Ni lattice; hence, the microscopic dipolar field is zero, $b_{\text{dip}}(\text{fcc}) = 0$.

In an hcp Co lattice, the dipolar field is also weak but is nonzero and has different values in crystallographically nonequivalent interstitial sites. If we direct the $z$ axis along the hexagonal axis, we have

$$\delta a_{xx}^h = \delta a_{yy}^h = \Delta/2, \quad \delta a_{zz}^h = -\Delta$$
$$\delta a_{xx}^t = \delta a_{yy}^t = -\Delta, \quad \delta a_{zz}^t = 2\Delta$$

in octahedral interstitial sites,

in tetrahedral interstitial sites,

where $\Delta \approx 0.1$. So, the dipolar field can be written as $b_{\text{dip}}(\text{hcp}) = \delta a_{ik}M_k$. Therefore, we have $b_{\text{dip}} \ll M$.

A more complicated picture is observed in a bcc Fe lattice, where the dipolar field is large and depends not only on the type of interstitial sites but also on the direction of the magnetization vector $M$. The components of the tensor $a_{ik}(\text{bcc})$ along the principal axes are [5, 6]

$$a_{xx}^h = a_{yy}^h = 1.165, \quad a_{zz}^h = 14.9$$
$$a_{xx}^t = a_{yy}^t = 5.707, \quad a_{zz}^t = 1.152$$

in octahedral interstitial sites,

in tetrahedral interstitial sites.

Hence, the local field acting on a muon in a bcc lattice is given by

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

The local field acting on a muon depends on the direction of the magnetization vector in the grain (see Eq. (5)). The polarization of the muons fraction that stop in the crystallographically ordered grains should be defined by averaging over all possible orientations of the principal crystallographic axes.

Let us represent the local field acting on a muon as the sum of the two components parallel and perpendicular to the film plane $b = b_\parallel + b_\perp$. Then, the polarization components are defined as

$$P_\perp = \langle b_\parallel e^{i\omega t} \rangle, \quad P_\parallel = \langle b_\perp \sin \omega t \rangle, \quad \omega = \gamma_{\mu} b / \gamma_{\mu} \sqrt{b_\parallel^2 + b_\perp^2}$$

The preexponential factors (the direction cosines) depend on whether the external field is perpendicular or parallel to the film plane.

3. Hierarchy of fields

To determine the local field acting on a muon implanted into a target, we need to find the microscopic field in the sample. At first, the hierarchy of fields in films of nanostructured ferromagnetic metals should be refined. As for a multidomain ferromagnet, in addition to the macroscopic fields in each grain $M, B, H$ and the external one with the respect to entire sample $B$, we need to introduce average macroscopic fields in the sample. These fields are
Thus, to determine the fields, we should determine the direction of the magnetization vector. If the external field lies in the film plane, we obtain up to an effective depolarization in accordance with Eq. (9). In the case of a strong external field, we have the local field acting on a muon (see Eqs. (12) and (16)) could be represented as a sum of two items, one of them is a constant and the other depends on a magnetization orientation.

\[ H_{\parallel} = B - 4\pi M_z, \quad H_{\perp} = Bpl = 4\pi (M_{pl} + M_{\parallel}). \]

4. Muon spin polarization description

We can see that the local field acting on a muon (see Eqs. (12) and (16)) could be represented as a sum of two items, one of them is a constant and the other depends on a magnetization orientation in a grain. The averaging of the muon spin polarization overall possible grain orientations leads up to an effective depolarization in accordance with Eq. (9). In the case of a strong external field, we have

\[ H_{\parallel} = B - 4\pi M_z, \quad H_{\perp} = Bpl = 4\pi (M_{pl} + M_{\parallel}) \]
field \( B \gg M \) the precession frequency of the muon spin polarization could be also written as a sum of two items, \( \omega = \omega_0 + \omega(\vartheta) \), where \( \vartheta \) is the angle between the grain magnetization and the external field. Only the transverse component of the muon spin polarization does not equal to zero, and it can be approximately written in the form

\[
P_\perp = b_\perp e^{-i\gamma \mu b t} = e^{-i\omega_0 t} e^{-i\omega(\vartheta) t} = e^{-i(\omega_0 + \Delta \omega) t} e^{-\sigma^2 t^2},
\]

Here, the frequency shift \( \Delta \omega \) and the depolarization rate \( \sigma \) are determined by both the grain magnetization value and the muon interstitial position. Analytical expressions for these important parameters were obtained in [8] in cases of rapidly diffusing and nondiffusing muons.

In the case of fast diffusing the muon spin polarization behaviour in fcc and bcc lattices is practically the same. Up to the quadratic terms of \( M/B \), the precession frequency \( \omega_0 \) and its shift \( \Delta \omega \) are given by

\[
b_0 = B - \frac{8\pi}{3} M + \frac{1}{2} \left( \frac{4\pi}{3} \right)^2 \frac{M^2}{B}, \quad \Delta \omega = \frac{1}{63} \frac{2\pi}{3} M \left( \frac{M}{B} \right)^2.
\]

The second moment \( \sigma^2_{\text{diff}} \propto (M/B)^2 \) is rather small and it is unlikely that it can be measured in experiments.

In the case of nondiffusing muons one can obtain more detail information. In a bcc lattice of ferromagnet belongs to the "easy axis" type (e.g. Fe) the precession frequency and its shift are determined by

\[
\omega_{0,\text{bcc}} = \gamma \mu (B - (2\pi + \frac{a_\perp}{2}) M), \quad \Delta \omega_{\text{bcc}} = -\frac{1}{3} \gamma \mu d M \left[ 1 - \left( 2d - \frac{41}{28} \beta \right) \frac{2M}{15B} \right],
\]

and the second moment

\[
\sigma^2_{\text{nd}} = \frac{7}{30} (\gamma \mu d M)^2.
\]

Here, \( a_\parallel \) and \( a_\perp \) are the components of the tensor \( a_{ik} \) defining the dipole field, \( d = (a_\parallel - a_\perp)/2 \), \( \beta \) is the magnetic anisotropy parameter.

The behaviour of the polarization of nondiffusing muons in uniaxial ferromagnets is similar to that presented above for cubic ferromagnets (19)–(20)

\[
\omega_{0,\text{hcp}} = \gamma \mu \left[ B - \left( \frac{8\pi}{3} - \frac{\Delta}{2} \right) M \right], \quad \Delta \omega_{\text{hcp}} = -\frac{1}{2} \gamma \mu M \Delta \left[ 1 - \left( \beta + \frac{3}{2} \Delta \right) \frac{4M}{5B} \right],
\]

\[
\sigma^2_{\text{nd}} = \frac{21}{40} (\gamma \mu M \Delta)^2.
\]

5. Conclusion

The formulas presented in this report show that the \( \mu \)SR-technique gives the opportunity to determine the magnetization in a grain by measuring the precession frequency and the depolarization rate and to reveal the existence of muon diffusion in the crystallographically ordered fraction of a sample. In the case of nondiffusing muons in a crystallographically ordered grain the precession frequency measurement allows one to determine the magnetic anisotropy constant, which is of great importance in the physics of the phenomenon under consideration. The measurement of the depolarization rate of the muon spins precessing at the frequency corresponding to the average magnetic field of the film allows determination of the characteristic scale of the magnetic field inhomogeneity in the disordered intergrain region. The ratio between the precession amplitudes of the two fractions of muons allows to determine the ratio of the
volumes of the paramagnetic and ferromagnetic phases of the sample. In addition, the local fields induced by nuclear magnetic moments can be taken into account separately using the well known approaches for normal metals [5]. We note that all stable isotopes of Fe and Ni, except $^{57}$Fe (2.21%) and $^{61}$Ni (1.25%), have zero spins (see e.g. [9]) and, hence, experiments with these widespread magnets are preferred.

Recent experiments [10]-[14] showed that low-energy muons (LE-$\mu$SR) give opportunities to study the magnetic properties and inhomogeneity of thin superconducting films. This LE-$\mu$SR technique has a scale resolution up to 10 nm and can be applied successfully to solve the fundamental problem of magnetic scale phase transitions in ferromagnets too. Simultaneous measurements by macroscopic methods (see e.g. [11]) would make it possible to obtain a complete information on the physics of the phase transitions in nanocrystalline ferromagnetic films.

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