Possibilities of μSR-technique for study of magnetization processes in nanocrystal films of ferromagnetic metals

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Abstract. It is shown that μSR-technique allows to study appearance of ferromagnetic ordering in nanocrystal films of ferromagnetic metals – so called "scale" phase transition. A relationship of a microscopic (local) field with macroscopic characteristics just as an external magnetic field, average magnetization and saturation magnetization is determined in a model of the nanocrystal film consisting of crystallographically ordered grains separated by disordered areas. Expressions for behaviour of a muon spin polarization ensemble in this kind of structures are obtained in cases of fast diffusing and nondiffusing muons. It is shown that experiments with "slow" positively charged muons allow to measure all parameters of this kind of structures and obtain important information for the study of phase transition physics.

1. Introduction

Magnetic nanocrystalline metals are of interest for μSR-technique since using positive muons and neutron difraction possess real possibilities to study their bulk properties[1]. Possible applications of μSR-technique for study nanocrystalline ferromagnetic materials were still mentioned at 2000-th [1]. Nevertheless, no serious experimental researches were carried out yet. There was emphasized that at least two problems in the fundamental physics of magnets could be solved for nanostructured ferromagnets. One of them is the problem of specific "scale" phase transitions, when an existence of a spontaneous magnetization depends both on temperature and sizes 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 differ from the respective characteristics of "ordinary" polycrystalline ferromagnets. Really, both theory and experiments show that a microcrystalline grain ($10^2 - 4 \cdot 10^2$ Å) should be a single domain. The characteristic thickness of domain walls in a bulk sample[2] is $d \sim 10 - 30$ 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 observed for macroscopic polycrystals. In our case we need to consider magnetized regions without domain grains in a

[1] Nanocrystalline metals are conventionally accepted to be polycrystals with grain sizes of 10–400Å.

[2] The characteristic thikness of domain walls depends on the ratio of exchange and magnetocrystalline anisotropy energy scale. The presented values are correct for ferromagnets under consideration.

nonmagnetic medium. Hence, the macroscopic field $B_{\rm dom}$ inside a crystallographically ordered grain and its dependence on the external magnetic field $\mathcal{B}$ should differ significantly from the properties of macroscopic samples.

Opportunities of the μSR-technique for studying ferromagnetic nanostructured thin films are represented in this report.

2. Behaviour of muon spin polarization

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

$$\mathcal{P}(t) = \mathbf{P}_{\rm cr}(t) + \mathbf{P}_{\rm nc}(t), \tag{1}$$

where $\mathbf{P}_{\rm cr}(t)$ and $\mathbf{P}_{\rm nc}(t)$ are the polarization of the fraction of muons stopped in grains and in intergrain regions, respectively.

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

$$P_i(t) = \mu_{ik}(t)P_k(0) \tag{2}$$

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

$$\mu_{ik}(t) = n_i n_k + (\delta_{ik} - n_i n_k)\cos\gamma_\mu b_\mu t + e_{ikl} n_l \sin\gamma_\mu b_\mu t \tag{3}$$

Here $\mathbf{b}_\mu$ is the local field exerted on the muon, $\gamma_\mu = 13.554$ kHz/G is the gyromagnetic ratio and $\mathbf{n} = \mathbf{b}_\mu/|\mathbf{b}_\mu|$ is the unit vector along the magnetic field.

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

$$\mathbf{b}_\mu = \mathbf{B} - \frac{8\pi}{3}\mathbf{M} + \mathbf{b}_{\rm dip} + \mathbf{B}_{\rm cont}, \tag{4}$$

where $\mathbf{B}_{\rm cont}$ is the contact field induced by electrons and $\mathbf{b}_{\rm dip}$ is the microscopic field induced by the oriented magnetic dipoles inside the Lorentz sphere. The contact field can always be written as $B_{i\,\rm cont} = K_{ik}B_k$. 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 ($\mathbf{M}_{\rm nc} = 0$), hence, the dipole fields $\mathbf{b}_{\rm dip}$ could be induced only by the disordered nuclear magnetic moments. These fields cause 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 integrain regions is controlled by the local magnetic field $\mathbf{b}_\mu = \langle\mathbf{B}\rangle + \delta\mathbf{b}$, where $\delta\mathbf{b}$ 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 to determine the average magnetic field in the film. In the case of nondiffusing muons a Gaussian exponent allows to determine the characteristic scale of the field inhomogeneity, $\sigma \sim \gamma_\mu\langle\delta\mathbf{b}^2\rangle/|\langle\mathbf{B}\rangle|$. In the case of diffusing muons depolarization rate depends

[3] For the simplicity, we can omit the Knight shift in what follows, although it can make an appreciable contribution in some cases.

on a diffusing rapid λ and in the fast diffusing limit becomes negligibly small. It is known that muon 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 dipole field is induced by the ordered electron magnetic moments. In this case, inside a Lorentz sphere, this field can always be written as [5, 6]

$$b_{i\,\mathrm{dip}} = -\frac{4\pi}{3}M_i + a_{ik}M_k, \tag{5}$$

where the tensor a_{ik} depends on the type of interstice at the center of which the field is determined. Calculations showed (see e.g.,[5, 6]) that $a_{ik} = \delta_{ik}4\pi/3$ in an fcc Ni lattice; hence, the microscopic dipole field is zero, $\mathbf{b}_{\mathrm{dip}}(fcc) = 0$.

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

$$\begin{array}{lll} \delta a^h_{xx} & = \delta a^h_{yy} = \Delta/2, \quad \delta a^h_{zz} = -\Delta & \text{in octahedral interstitice,} \\ \delta a^t_{xx} & = \delta a^t_{yy} = -\Delta, \quad \delta a^t_{zz} = 2\Delta & \text{in tetrahedral interstice,} \end{array} \tag{6}$$

where $\Delta \approx 0.1$. So, the dipole field can be written as $\mathbf{b}_{\mathrm{dip}}(hcc) = \delta a_{ik}M_k$. Therefore, we can decide the dipole field $b_{\mathrm{dip}} \ll M$.

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

$$\begin{array}{lll} a^h_{xx} & = a^h_{yy} = -1.165, \quad a^h_{zz} = 14.9 & \text{in octahedral interstice,} \\ a^t_{xx} & = a^t_{yy} = \quad 5.707, \quad a^t_{zz} = 1.152 & \text{in tetrahedral interstice.} \end{array} \tag{7}$$

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

$$b_{\mu\,i}(bcc) = B_i - 4\pi M_i + a_{ik}(bcc)M_k. \tag{8}$$

The local field acting on a muon depends on the direction of the magnetization vector in the grain (see Eq. (5)), the polarization fraction of muons 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 $\mathbf{b} = \mathbf{b}_{\parallel} + \mathbf{b}_{\perp}$. Than, the polarization components are defined as

$$\mathcal{P}_{\perp} = \langle \frac{b_{\parallel}}{b} \mathrm{e}^{\imath\omega t} \rangle, \quad \mathcal{P}_{\parallel} = \langle \frac{b_{\perp}}{b} \sin \omega t \rangle, \qquad \omega = \gamma_\mu b = \gamma_\mu \sqrt{\mathbf{b}^2_{\parallel} + \mathbf{b}^2_{\perp}} \tag{9}$$

The preexponential factors (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 macroscopic fields in *each* grain $\mathbf{M}$, $\mathbf{B}$, $\mathbf{H}$ and the external with the respect to *entire* sample $\boldsymbol{\mathcal{B}}$ we need to introduce average macroscopic fields in the sample. These fields are the results of an averaging of all mentioned above macroscopic fields and the magnetization over all grains and intergrain regions, $\langle \mathbf{B} \rangle$, $\langle \mathbf{H} \rangle$ $\langle \mathbf{M} \rangle$. The last parameters are determined in macroscopic experiments. The same parameters determine the field acting on a muon in a disordered intergrain region of the sample.

Let us denote the demagnetization factors of the film as N_{ik} and write the relations between the average fields and the external field in the form

$$\mathcal{B}_i = \langle H_i \rangle + 4\pi N_{ik} \langle M_k \rangle, \quad \langle \mathbf{B} \rangle = \langle \mathbf{H} \rangle + 4\pi \langle \mathbf{M} \rangle. \tag{10}$$

Let the z axis of the coordinate system associated with the film be perpendicular to the film plane. Since, by definition, the film thickness d is much smaller than its linear dimensions L, we have $N_{zz} \equiv N_\perp \approx 1$ and all other components are negligibly small.

If the external field is perpendicular to the film plane $\boldsymbol{\mathcal{B}} \| z$, we have

$$\mathcal{B}_z \equiv \mathcal{B} = \langle H_z \rangle + 4\pi \langle M_z \rangle, \quad \langle H_{\rm pl} \rangle \approx 0, \tag{11}$$

where $H_{\rm pl}$ are the vector $\mathbf{H}$ components lying in the film plane. From the relation (10) we obtain

$$\langle B_z \rangle = \mathcal{B}, \quad \langle B_{\rm pl} \rangle \approx 4\pi \langle M_{\rm pl} \rangle. \tag{12}$$

If the external field lies in the film plane $\boldsymbol{\mathcal{B}} \perp z$, we have

$$\langle H_z \rangle + 4\pi \langle M_z \rangle = 0, \quad \langle H_z \rangle = -4\pi \langle M_z \rangle, \quad \langle H_{\rm pl} \rangle \approx \mathcal{B} \tag{13}$$

and the average induction is given by

$$\langle B_z \rangle = 4\pi \langle M_z \rangle, \quad \langle B_{\rm pl} \rangle = \mathcal{B} + 4\pi \langle M_{\rm pl} \rangle. \tag{14}$$

In the accepted model of the film a nanocrystal grain is a single-domain particle. Therefore, in contrast with the situation in an ordinary ferromagnet, each grain has no domain wall and is in both the external (homogeneous) field $\boldsymbol{\mathcal{B}}$ and the field induced by the magnetized grains of the film.

For each grain (in the form of ellipsoid) we can write a relation similar to Eq. (10), $\langle B_i \rangle = H_i + 4\pi n_{ik} M_k$, where n_{ik} are the grain demagnetization factors. Analytical expressions could be obtained in the approximation of oblate ellipsoids, when $n_\| = n_{zz} \approx 1, \quad n_\perp = 1 - n_\| \ll 1$.

If the external field is perpendicular to the film plane, Eqs. (12) give

$$\langle B_z \rangle \approx \mathcal{B}, \qquad H_z = \mathcal{B} - 4\pi M_z \qquad H_{\rm pl} = \langle B_{\rm pl} \rangle = 4\pi \langle M_{\rm pl} \rangle, \quad B_{\rm pl} = 4\pi \big(\langle M_{\rm pl} \rangle + M_{\rm pl} \big). \tag{15}$$

If the external field lies in the film plane, we obtain

$$H_z = -4\pi M_z, \quad H_{\rm pl} \approx \langle B_{\rm pl} \rangle = \mathcal{B} + 4\pi \langle M_{\rm pl} \rangle \qquad B_z = 0, \quad B_{\rm pl} = \mathcal{B} + 4\pi \big(\langle M_{\rm pl} \rangle + M_{\rm pl} \big). \tag{16}$$

Thus, to determine the fields, we should determine the direction of the magnetization vector $\mathbf{M}$ in each grain and $\langle \mathbf{M} \rangle$ in the film. The local field was obtained in [8] in the approximation when the ordered grains are considered as oblate ellipsoids.

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. An averaging of a muon spin polarization overall possible grain orientations leads up to an effective depolarization in accordance with Eq. (9). In a case of strong external field ($\mathcal{B} \gg M$) a precession frequency of a muon spin polarization could be written as a sum of two items too, $\omega = \omega_0 + \omega(\vartheta)$, where ϑ is the angle between the grain magnetization and the external field. At this approach only the transverse component of a muon spin polarization does not equal to zero, and it can be approximately written in a form

$$\mathcal{P}_\perp = \langle \frac{b_z}{b} \mathrm{e}^{-\imath\gamma_\mu bt} \rangle \approx \mathrm{e}^{-\imath\omega_0 t} \langle \mathrm{e}^{-\imath\omega(\vartheta)t} \rangle = \mathrm{e}^{-\imath(\omega_0+\Delta\omega)t} \mathrm{e}^{-\sigma^2 t^2}, \qquad (17)$$

Here the frequency shift $\Delta\omega$ and the depolarization rate σ 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 a muon spin polarization behaviour in fcc and bcc lattices is practically the same. To within terms quadratic in $M/\mathcal{B}$ the precession frequency ω_0 and its shift $\Delta\omega$ are given by

$$b_0 = \mathcal{B} - \frac{8\pi}{3} M + \frac{1}{2} \left(\frac{4\pi}{3} \right)^2 \frac{M^2}{\mathcal{B}}, \quad \Delta\omega = \gamma_\mu \frac{1}{63} \frac{2\pi}{3} M \left(\frac{M}{\mathcal{B}} \right)^2. \qquad (18)$$

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

In the case of nondiffusing muons one can obtain the 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

$$\omega_0^{bcc} = \gamma_\mu (\mathcal{B} - (2\pi + \frac{a_\perp}{2}) M), \quad \Delta\omega^{bcc} = -\frac{1}{3} \gamma_\mu d\, M \left[1 - \left(2d - \frac{41}{28} \beta \right) \frac{2M}{15\mathcal{B}} \right], \qquad (19)$$

and the second moment

$$\sigma^2_{\mathrm{nd}} = \frac{7}{30} (\gamma_\mu d\, M)^2 . \qquad (20)$$

Here $a_\parallel$ and $a_\perp$ are the components of the tensor a_{ik} defining the dipole field, $d = (a_\parallel - a_\perp)/2$, β 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)

$$\omega_0^{hcp} = \gamma_\mu \left[\mathcal{B} - \left(\frac{8\pi}{3} - \frac{\Delta}{2} \right) M \right], \quad \Delta\omega^{hcp} = -\frac{1}{2} \gamma_\mu M \Delta \left[1 - (\beta + \frac{3}{2}\Delta) \frac{4M}{5\mathcal{B}} \right],$$
$$\sigma^2_{\mathrm{nd}} = \frac{21}{40} (\gamma_\mu M \Delta)^2 . \qquad (21)$$

5. Conclusion

The formulas presented in this report show that the μSR-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.

Last experiments [10]-[14] showed that low-energy muons (LE-μSR) give opportunities to study magnetic properties and inhomogeneity of thin superconducting films. This LE-μSR 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 complete information on the physics phase transitions in nanocrystalline ferromagnetic films.

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