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About the influence of nuclear interactions on longitudinal magnetic relaxation of ^{57}Fe nuclei in YIG films

S.N.Polulyakh, N.A.Sergeev*

Taurida National University, 95007 Simferopol, Ukraine *Institute of Physics, University of Szczecin, 70-451 Szczecin, Poland

The effect of impurity ions on nuclear magnetic relaxation has been considered. It is supposed that an impurity ion introduces an excess electron (or a hole). As a result, magnetic ions with strong spin-orbital interaction appear and magnetization direction distribution becomes inhomogeneous. Thermally activated jumps of the excess electron from one magnetic ion to another generate fluctuations in the magnetization direction. The interaction between the fluctuating magnetization and magnetic nuclei results in the appearance of regions of fast nuclear magnetic relaxation. Spin diffusion contributes to the longitudinal relaxation rate also for nuclei being outside the impurity regions. Taking into account the spin diffusion dependence on nuclear interactions, we have derive the dependence of longitudinal relaxation rate on nuclear interactions, what was observed experimentally in [1]. An assumption about unintentional technological impurities is sufficient to explain the experimental data.

Рассматривается влияние примесных ионов на процессы ядерной магнитной релаксации. Предполагается, что примесный ион привносит избыточный электрон (или дырку). В результате появляются магнитные ионы с сильной спин-орбитальной связью и распределение направлений электронной намагниченности становится неоднородным. Термически активированные скачки избыточного электрона с одного магнитного иона на другой порождают флуктуации в направлении намагниченности. Взаимодействие между флуктуирующей намагниченностью и магнитными ядрами приводит к появлению областей с быстрой ядерной магнитной релаксацией. Благодаря спиновой диффузии возрастает скорость продольной релаксации и для ядер, находящихся вне примесных областей. Принимая во внимание зависимость интенсивности спиновой диффузии от ядерно-ядерных взаимодействий, получена зависимость скорости продольной релаксации от межядерных взаимодействий, что наблюдалось экспериментально в работе [1]. Предположение о наличии неконтролируемых технологических примесей является достаточным для объяснения экспериментальных результатов.

The Nuclear Magnetic Resonance (NMR) is a powerful method to study magnetic materials due to its microscopic nature. As a rule, it is just the NMR spectra, which reflect the hyperfine field distribution, that are the subject of NMR investigations. The opposite case takes place in the analysis of nuclear magnetic relaxation data. The strong sensitivity of relaxation parameters to impurities, even to the unintentional technological ones, is among possible difficulties in that way.

The nuclear magnetic relaxation is subdivided into transverse and longitudinal components. It is well known [2] that the nuclear interaction influences the transverse relaxation only, if the rigid lattice model is considered. On the other hand, the influence of abundant magnetic nuclei on the longitudinal relaxation has been observed experimentally in yttrium-iron garnet (YIG) films at 77 K. Discussion of the mechanisms responsible for this phenomenon is the matter of this work.

We have developed a concept proposed in [3–5]. The main idea is the assumption on the influence of unintentional technological non-magnetic impurities which cause a redistribution of ion charges. As a result, the ions of alternative charge appear in magnetic lattice of the sample. The most difficult point of the approach presented in [3–5] is the assumption on energy levels of such magnetic ions: it is supposed, the ion has magnetic levels at a distance consistent with the NMR frequency (of order of tens MHz for 3*d* ions). In contrast, we have developed the concept of fluctuations in magnetization orientation, proposed before for the analysis of relaxation data [6]. We have shown here, that the reason for these fluctuations is the thermally activated migration of excess (or missing) charge introduced by the impurity ion. The impurity regions for the impurity ions are regions of short longitudinal relaxation rate, so that the assumption on unintentional technological impurities and spin diffusion is sufficient to explain the experiments described in [1].

The YIG free of impurities contains magnetic ions of one type only: these are Fe^{3+} ions. The impurity ions, which contribute an excess electron (or a hole) stimulate the appearance of Fe^{2+} or Fe^{4+} ions. The differently charged magnetic ions differ also in magnetic moment, exchange interactions and more strong spin-orbital interaction. To analyze the influence of differently charged ions on magnetic properties of YIG, we have built a simple computer simulation model. The model deals with magnetic ions only. The classical magnetic moments μ have been placed into octahedral and tetrahedral positions of the garnet lattice. The negative exchange interactions of Heisenberg type between nearest ions have been taken into account. The zero point has been chosen at an octahedral position and magnetic moment at this point has been set to be $(1 - 1/5)\mu$. The ion at zero point simulates oppositely charged magnetic ion. To simulate the initial state, we have directed magnetic moments of all ions collinearly to the <111> lattice direction (easy magnetization direction in YIG). The model deals with ions being at distances from the center not exceeding $5a$ (a — YIG lattice constant, total 20993 ions).

The effective magnetic field of exchange interactions applied to the ion has been calculated as $\mathbf{B}_{ex} = J\sum \mu_j$. Here, J is the exchange interaction constant, the summation is assumed over the nearest neighbors only. For the boundary ions, the existence of "imagine" ions was supposed and direction of magnetic moments for these ions was strongly associated with <111> direction. To simulate the spin-orbit interactions, we have introduced an additional effective magnetic field $\mathbf{B}_{SL}$ directed along <111> direction and applied to the oppositely charged ion only. To build the equilibrium state, we sequentially select ions and direct the magnetic moment of ion strictly along the effective magnetic field. The energy of interaction between magnetic moment and effective field has been used as a criterion for the iterations. The calculated deviations in directions of magnetic moments from the initial <111> one as a function of ion distance is given in Fig. 1 for the case $B_{LS} = 10^{-3}J\mu$. We have assumed the spin-orbit interactions to result in deviations of magnetic moment orientation of oppositely charged ion from equilibrium direction for the crystal. Due to the exchange interactions, this deviation is transferred to other magnetic ions, so that the weak spin-orbit interaction (three decimal orders weaker than the exchange interaction) can provide deviation about 1° in the size range of about several unit cells. Deviations of such order have been used to explain the experiments in [6].

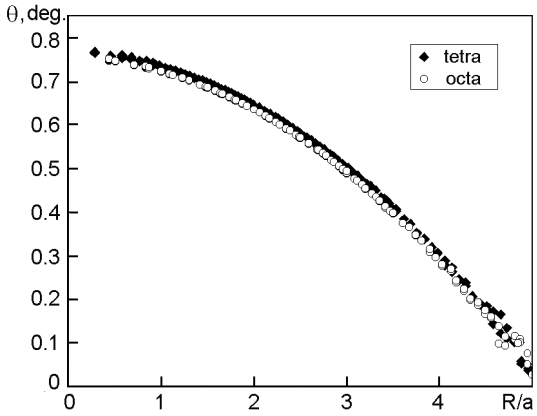

Fig. 1. Calculated dependence of deviation in magnetization direction on the distance from impurity ion.

Special experiments performed on YIG films doped with Si have shown that the excess electron (or a hole) migrates from one iron ion to another [7]. The frequency of those migrations exceeds the NMR frequency [7]. Taking into account different

local axes for different lattice positions, we conclude that the migration of excess electron (or a hole) generates fluctuations in the magnetization direction.

When analyzing nuclear magnetic relaxation, let us follow the approach of [3–5]. We suppose the following relaxation mechanisms: (i) Direct fluctuations of local magnetic fields caused by fluctuations in magnetization direction and hyperfine field fluctuations caused by change in ion charges; (ii) Interaction involving virtual magnons between magnetic nuclei (Suhl-Nakamura interactions [2]) and between magnetic nuclei and fluctuated magnetization. The last mechanism is important if the magnetization fluctuations have non-zero spectral density at NMR frequency.

As a result, we can subdivide all the nuclei into two types, which are different in relaxation rate. The uclei of fast magnetic relaxation are localized within the size range of R_{SN} near impure ion. Here, R_{SN} is the distance of Suhl-Nakamura interactions. The fast magnetic relaxation for the first-type nuclei is caused by magnetization fluctuations, which take place for the magnetization value as well as for magnetization direction and are caused by thermally stimulated migrations of excess charge introduced by impurities. All other nuclei in the sample are nuclei of slow magnetic relaxation.

If the magnetic relaxation in a sample is inhomogeneous, the spin diffusion takes place. Following to [4], we can write the nuclear diffusion equation as

$$\frac{\partial m_z}{\partial t} = D\Delta m_z - \frac{m_z - m_0}{T_{1f}}.$$

Here m is nuclear magnetization dependent both on time and position; T_{1f}, the longitudinal relaxation time for the fast-relaxing nuclei; $D = R_{SN}^2/3T_2$, the diffusion parameter; and T_2, the transverse relaxation time caused by interaction between nuclei. Assuming average distance R between the fast magnetic relaxation centers (distance between impurities) is large as compared to R_{SN}, we obtain for longitudinal relaxation time outside the impurity centers

$$T_1^{-1} = T_2^{-1}\frac{R_{SN}^3}{R^3}.$$

The relationship between transverse and longitudinal relaxation is known from experiment for the sample enriched in magnetic nuclei: $T_1^{-1} = T_2^{-1} \approx 0.01$ [1]. The distance of Suhl-Nakamura interactions can be estimated as $R_{SN} \sim (10^2 - 10^3)a$, where a is the lattice constant [2]. As a result, we can estimate the average distance between impurity centers as $R \approx 0.5(10^3 \div 10^4)a$.

Thus, the average distance between impurity centers, as derived from experimental values for relaxation times, evidences a very low impurity concentration. The dependence of longitudinal relaxation time on nuclear abundance is caused by spin diffusion and fast nuclear relaxation near impurities. In contrast to [3–5], we do not need to have for energy levels of impurity ions the distances of the NMR frequency order. The magnetization fluctuations at NMR frequency appear as a result of thermal migrations of excess electron (or hole), which appears in lattice due to the impurity ion.

References

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Про вплив ядерних взаємодій на поздовжню магнітну релаксацію ^{57}Fe у плівках залізо-ітрієвого гранату

С.Н.Полулях, Н.А.Сергєєв

Розглядається вплив домішок на процеси ядерної магнітної релаксації. Робиться припущення, що іон домішки надає надлишковий електрон (або дірку). Внаслідок цього з'являються магнітні іони із сильним спин-орбітальним зв'язком і розподіл напрямку електронної намагніченості стає неоднорідним. Термічно активовані стрибки надлишкового електрона з одного магнітного іона на інший породжують флуктуації в напрямку намагніченості. Взаємодія між флуктуаціями намагніченості та магнітними ядрами призводить до появи регіонів зі швидкою ядерною магнітною релаксацією. Завдяки спіновій дифузії зростає швидкість подовжньої релаксації і для ядер, що знаходяться поза регіонами домішок. Приймаючи до уваги залежність інтенсивності спінової дифузії від ядерно-ядерних взаємодій, отримано залежність швидкості подовжньої релаксації від взаємодій між ядрами, що спостерігалося експериментально в роботі [1]. Припущення про наявність неконтрольованих технологічних домішок є достатнім для пояснення експериментальних результатів.