Fluctuating spin density waves revealed by perturbed angular correlation spectroscopy

C. L'abbé
Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439

J. Meersschaut[a)]
Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439 and Instituut Kern- en Stralingsfysica, K.U. Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

M. Rots
Instituut Kern- en Stralingsfysica, K.U. Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

S. D. Bader
Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Illinois 60439

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We present a theory to analyze pertubed angular correlation spectra in the framework of dynamical processes. It can be applied to study superparamagnetic properties of magnetic nanostructures, and was used recently to identify dynamical fluctuations in spin-density-wave ordered Cr. We showed that the spin fluctuations give rise to the biquadratic coupling in Fe/Cr(100) multilayers [J. Meersschaut, C. L'abbé, M. Rots, and S. D. Bader, Phys. Rev. Lett. **87** 107201 (2001)]. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558652]

I. INTRODUCTION

Dynamical processes and relaxation phenomena play an important role in various branches of solid-state physics. Examples are the diffusion of impurities and vacancies in metals,[1] the roughening of high-index metal surfaces,[2] and the tunneling of positive particles in superconductors.[3] It is expected that relaxation phenomena will gain even more interest as researchers are further reducing the dimensions of magnetic media to achieve higher recording densities, thereby reaching the superparamagnetic limit.[4] However, only a few techniques that can probe time-dependent phenomena are at hand. Electromagnetic spectroscopies can measure correlation times between 10^2 and 10^{-6} s. Neutron diffraction, on the other hand, can probe correlation times of 10^{-9} s down to 10^{-14} s. Hyperfine interaction techniques can provide information on the dynamical behavior in the intermediate time-scale, and, moreover, are easily applied to nanostructures.[5–7] In Ref. 8 we used perturbed angular correlation (PAC) spectroscopy to study Fe/Cr(100) superlattices, and discovered dynamical spin fluctuations in the Cr spacer layers. Here, we will present in more detail the procedure that was followed for the analysis.

II. FLUCTUATING HYPERFINE FIELDS IN PERTURBED ANGULAR CORRELATION SPECTROSCOPY

In a time-differential perturbed angular correlation experiment, one studies the correlation between two successive photons emitted in a nuclear decay cascade. At time $t=0$ the nucleus decays from its initial excited state to an intermediate state $|Im\rangle$ via emission of a first photon. The intermediate state will be polarized leading to an unequal population of the angular momentum states. After some time t, this intermediate state decays to the final state through emission of a second photon. The presence of hyperfine fields will cause a reorientation of the nuclear moment during the lifetime of the intermediate state. As a result, the angular distribution pattern of the second γ ray is not only correlated to the emission direction of the first γ ray, but in addition, it is strongly modulated by the hyperfine fields acting on the intermediate nuclear state. Measuring the angular correlation as a function of the time elapsed between the two γ emissions provides essential information on the hyperfine interactions.

The time evolution of the intermediate state is described by the perturbation factors $G_{kk'}^{NN'}(t)$. If the hyperfine interactions are static, an expression for the perturbation factors can easily be derived.[9] If, however, the hyperfine interactions are not static but fluctuate in time, an expression for the perturbation factors can be obtained by following the procedure described later. We use Winkler and Gerdau's quantum mechanical theory to describe the influence of fluctuating hyperfine fields in PAC.[10] It is based on Blume's stochastical model[11] that uses the Liouville operator formalism to construct a nonhermitian complex supermatrix that depends on stochastical and quantum mechanical variables.

Consider a system whose Hamiltonian jumps at random as a function of time between a finite number of possible forms $H_\alpha, H_\beta, \ldots, H_x$. All individual Hamiltonians are independent of time (i.e., each individual Hamiltonian describes a static hyperfine interaction), but, at certain moments the interaction can change from one state to another. We adopt the continuous-time random walk strategy[12] and divide the time axis in discrete intervals of Δt. At any multiple of Δt the system can change to another hyperfine interaction. For realistic calculations, Δt should be chosen infini-

[a)]Electronic mail: johan.meersschaut@fys.kuleuven.ac.be

tesimally small. The "jump" probability is given by the stochastic matrix W, whose elements $W_{\beta\alpha}$ give the probability to jump from H_α to H_β after a time interval Δt. Assuming a stationary Markov process, i.e., the probability for a hyperfine field jump is independent of the history of the system and independent of the time at which this jump occurs, then the probability matrix W may be cast into the following form: $W_{\beta\alpha}=\delta_{\beta\alpha}+\lambda_{\beta\alpha}\Delta t$. The quantities $\lambda_{\alpha\alpha}=-1/\tau_\alpha$ are related to the mean lifetime τ_α of the stochastic state H_α, whereas the off-diagonal elements $\lambda_{\beta\alpha}$ are the transition probabilities per unit time. Since the probability matrix obeys the normalization property, it satisfies the relations: $\Sigma_\beta W_{\beta\alpha}=1$ and $\Sigma_\beta\lambda_{\beta\alpha}=0$.

One can now construct a supermatrix A which is a Liouville operator labeled by three indices: one stochastic index referring to the possible states of the hyperfine interaction, and two quantum mechanical indices corresponding to the different nuclear levels in the intermediate state. This yields a matrix of rank $x\cdot(2I+1)^2$. The elements of A are defined through the following relation:

$$\begin{aligned}(\beta m' M'|A|\alpha m M) = &-i\,\delta_{\alpha\beta}\delta_{MM'}\langle m'|H_\alpha|m\rangle/\hbar \\ &+i\,\delta_{\alpha\beta}\delta_{mm'}\langle M|H_\alpha|M'\rangle/\hbar \\ &+\delta_{mm'}\delta_{MM'}\lambda_{\beta\alpha}. \qquad (1)\end{aligned}$$

The time evolution of the intermediate state due to the presence of fluctuating hyperfine interactions is given by e^{At}. In order to obtain a useful expression for the perturbation factors, one needs to diagonalize the matrix A. If L_Z and R_Z are the left and right eigenvectors and Ω_Z the corresponding eigenvalues, then the perturbation factors are given by[10]

$$\begin{aligned}G_{kk'}^{NN'}(t) = &\sum_{mm'}(-1)^{2I+m+m'}\sqrt{(2k+1)(2k'+1)} \\ &\times\begin{pmatrix} I & I & k \\ M & -m & N\end{pmatrix}\begin{pmatrix} I & I & k' \\ M' & -m' & N'\end{pmatrix} \\ &\times\sum_{\alpha\beta Z} p_\alpha(\beta m' M'|L_Z)(R_Z|\alpha m M)e^{\Omega_Z t}. \qquad (2)\end{aligned}$$

Since the matrix A in general is non-Hermitian, the eigenvalues Ω_Z will be complex leading to a dampening of the PAC spectra. In Eq. (2), the p_α are the *a priori* probabilities for finding the nuclear environment at $t=0$ in the stochastic state $|\alpha\rangle$. The large brackets are the 3J symbols as defined in Ref. 9.

Equation (2) gives an expression for the perturbation factors used to calculate the angular correlation pattern. In a PAC experiment one usually takes a combination of several spectra recorded in different geometries (90° and 180° between the emission directions of the two successive photons). By properly combining the spectra, a time-dependent anisotropy function $R(t)$ is obtained (see Ref. 13). The anisotropy function contains all essential information on the hyperfine interactions and is more closely related to the experiment than the perturbation factors are. In the following we will calculate the $R(t)$ function for different lifetimes of the stochastic states in case of ^{111}Cd probe nuclei ($I=5/2$) embedded in Cr.

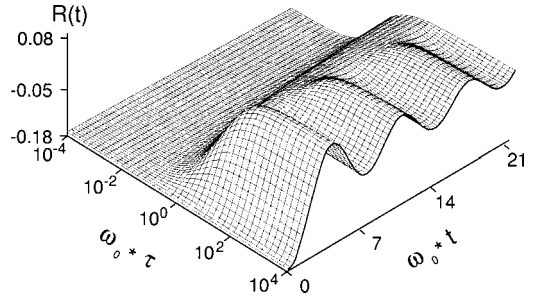

FIG. 1. PAC spectrum calculated for magnetic hyperfine fields whose magnitudes follow an Overhauser distribution with cutoff frequency ω_0 and whose directions fluctuate among two opposite states in the detector plane. Taken from Ref. 8.

III. DYNAMICAL SPIN FLUCTUATIONS IN Cr

We used perturbed angular correlation spectroscopy to study the incommensurate spin density waves in Cr.[14] The magnetic hyperfine field B is connected to the magnetization and shows a sinusoidal variation in space: $B(r)=B_0\sin(r)$. Correspondingly, the probability distribution for the field B is given by $P(B)=2/\pi(1/\sqrt{B_0^2-B^2})$. Or, equivalently, in terms of the Larmor frequencies ω_B: $P(\omega_B)=(2/\pi)\times(1/\sqrt{\omega_0^2-\omega_B^2})$ with ω_0 the cutoff frequency. $P(\omega_B)$ is known as the Overhauser distribution. In our calculations, the Overhauser distribution was sampled by 1000 ω_B points.

We have calulated the $R(t)$ function in case of an Overhauser distribution when the nuclei are subject to magnetic hyperfine fields that can fluctuate among two opposite directions, 45° in between the detectors. We assumed that the two hyperfine interactions are equally likely at time $t=0$ so that $p_\alpha=1/x$ (x being the number of possible hyperfine interactions; $x=2$ in our case), and that the probability per unit time to jump from one hyperfine interaction to another is equal for both stochastic states, i.e., there is no preferred state. Then, the transition probability matrix elements are determined via: $\lambda_{\alpha\beta}=-1/\tau$ for $\alpha=\beta$ and $\lambda_{\alpha\beta}=1/[(x-1)\tau]$ for $\alpha\neq\beta$, where τ is the mean lifetime of the stochastic states.

Simulations of the PAC spectra are shown in Fig. 1. In the slow fluctuation limit, we find the Bessel function of zeroth order, corresponding to the static case. In the intermediate range where the lifetime of the stochastic states is comparable to the characteristic frequency ω_0, the spectrum is strongly damped. In the very fast fluctuation limit, the nucleus sees the time-averaged field, which in this case is zero. Therefore, the fast fluctuation limit results in a paramagnetic-like spectrum. In the next section, we explain how these simulations were used to understand the biquadratic coupling in Fe/Cr(100) multilayers.

IV. BIQUADRATIC COUPLING IN Fe/Cr(100)

More than a decade ago, detailed investigations on the Fe/Cr/Fe layered system revealed the existence of biquadratic interlayer coupling.[15] The conventional theories for interlayer coupling, capable of explaining the long and short period oscillations in the interlayer coupling strength, are not able to explain the perpendicular alignment of neighboring Fe layers. Only a few theories were introduced that may

account for the existence of biquadratic interlayer coupling. In the fluctuation mechanism,[16] the biquadratic coupling is due to frustration of the bilinear exchange coupling. This frustration is attributed to one monolayer high steps at the interfaces. The fluctuation mechanism accounts for experimental data on biquadratic coupling obtained on the Fe/Ag/Fe and Fe/Cu/Fe systems. On the other hand, the strong temperature dependence of the biquadratic coupling observed in the Fe/Al/Fe or the Fe/FeSi/Fe system is most easily explained by the loose spin model.[17] In its original concept, the loose spins are either independent magnetic impurities or magnetic atoms located at the surface of the ferromagnetic metal.

In the case of the Fe/Cr/Fe system, it was concluded that none of the proposed mechanisms can account quantitatively for the experimental data.[18] The interlayer coupling in the Fe/Cr/Fe system is determined by the magnetic structure of the Cr spacer, which, in turn, may depend on its thickness,[19] on its crystalline structure,[20] or on the Fe/Cr interface.[21] In order to investigate the magnetic ordering in the Cr spacer layers when biquadratic coupling is present, we studied Fe/Cr multilayers with thick Cr spacers, where the biquadratic coupling was found to dominate.

The Fe/Cr multilayer studied in Ref. 8 was $[\mathrm{Fe}\ (1.7\ \mathrm{nm})/\mathrm{Cr}\ (8.4\ \mathrm{nm})]_{10}$. At 77 K, i.e., well below the blocking temperature, the biquadratic interlayer coupling was suppressed. The interlayer coupling strength was maximum at 225 K and decreased with further increasing temperature. We used perturbed angular correlation spectroscopy to probe the local magnetic properties of the Cr spacer layers. PAC spectra were recorded at three different temperatures. The spectrum recorded at 77 K was analyzed assuming a static magnetic spin-density-wave ordering in the Cr. The PAC spectra taken at 225 and 300 K were analyzed using the stochastical model for fluctuating hyperfine interactions that was described earlier. The characteristic lifetimes for the direction of the Cr moments were $\tau = 2.2$ ns and $\tau = 0.6$ ns, respectively. Through comparison with magnetization data, it became clear that the fluctuating Cr moments give rise to the biquadratic interlayer coupling. Furthermore, we explained how the loose spin model may be reinterpreted in the present context.

It is remarkable that the phase transition in Cr thin films from the low-temperature spin-density-wave ordering to the high-temperature paramagnetic state occurs through a superparamagnetic-like behavior. This gradual transition can be recognized in the resistivity measurements as a smearing out of the anomaly at the Néel temperature. It was observed that the anomaly in the resistivity is also smeared out for strained Cr or Cr alloys.[14,22] It is therefore tempting to believe that the observed phenomenon is of broader importance, and that spin fluctuations may occur in some of these cases as well.

To summarize, we have applied a stochastical theory for fluctuating hyperfine interactions in PAC spectroscopy in order to explain experimental data on Fe/Cr(100) superlattices. The analysis revealed the existence of fluctuating magnetic moments in the Cr layers. The interlayer coupling is strongly correlated with the temperature dependence of the fluctuation rates. Therefore, loose Cr spins are believed to be at the origin of biquadratic interlayer coupling.

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