

Non-cyclic geometric phase of nuclear quadrupole resonance signals of powdered samples

O. Glotova^{a,*}, N. Ponamareva^b, N. Sinyavsky^b, B. Nogaj^a

^a Department of Physics, Adam Mickiewicz University, Umultowska 85, 61-614 Poznań, Poland

^b Baltic State Academy, Molodiozhnaya str. 6, 236029 Kaliningrad, Russia

ARTICLE INFO

Article history:

Received 16 October 2010

Received in revised form

26 February 2011

Available online 10 March 2011

Keywords:

Nuclear quadrupole resonance

Geometric phase

Nutation

Asymmetry parameter

ABSTRACT

The non-cyclic geometric phase of ^{14}N and ^{35}Cl NQR signals induced by the character of trajectory of nuclear magnetization motion upon pulse r.f. excitation of powdered samples is studied. Analytical expressions for the geometric phases of NQR signals of the nuclei of spins $I=1$ and $3/2$ upon nuclear magnetization rotation induced by means of r.f. pulses with frequency detuned from the resonance and for any impulse duration for a separate crystallite are obtained. It is shown that the geometric phase recorded for the signal from a powdered sample at $\Delta\omega=0$ can be different from zero and can oscillate upon changes in duration of the r.f. excitation pulse. An alternative variant of the nutation experiment aimed at obtaining the asymmetry parameter η from locations of frequency singularities in the nutation phase spectrum for nuclei of spin $I=3/2$ in powder substances is proposed.

© 2011 Elsevier Inc. All rights reserved.

1. Introduction

Since the publication of the well-known Berry's work [1], the concept of geometric phase has been significantly extended and applied in various branches of physics, including radiospectroscopy. There are many theoretical publications devoted to the Berry phase in NQR and NMR.

The first experiment of pure NQR illustrating the Berry phase was carried out by Tycko [2], who used a monocrystal of sodium chlorate, in which the quadrupole nuclei of chlorine are oriented along the axis of crystal symmetry, which being also the axis of quantization. At crystal rotation about any axis, the two spin states acquire geometric phases, which increase in time leading to changes in the frequencies of transitions between these two states. This change was manifested as a shift of NQR frequencies of the nuclei and measured by means of standard phase-sensitive methods of detection.

The non-adiabatic geometric phase was observed by Suter et al. [3] using the NMR method. They studied a system of coupled protons, i.e. a quantum system with the total spin $I=1$. In their experiment the subspace with two levels was exposed to cyclic evolution induced by application of a time-dependent magnetic field. The geometric phase was measured on the basis of interference between these two states and the third, undisturbed level.

Zee [4] decided to perform the Tycko's experiment applying the rotation not around one axis but around two axes and proved

the appearance of non-Abelian Berry phase. A similar experiment was repeated by Zwanzinger and Koenig [5]. Zwanzinger's [6] work describes the changes in the geometric component of probability of transition in the system studied by NMR with two levels at a frequency sweep of r.f. irradiation. Furman and Kadzhaya [7] have shown theoretically that in a non-resonance case the shift of NMR frequency consists of two parts: the Bloch–Sigert shift and a shift of the Berry effect. Appelt et al. [8], using optical detection of ^{131}Xe NQR, extended the Tycko's experiment into non-adiabatic area, and for analysis of the Berry phase they applied the rotating frame of reference. The Berry dephase as a result of diffusion was investigated by Jones and Pines [9], applying optical detection of NQR in gas ^{131}Xe . The nontrivial mixing of spin states at a rotation of the system studied by NQR around the two axes and formation of a non-Abelian Berry phase is shown in Asakura's work [10]. Gopinath and Kumar [11] proposed the use of the Berry phase in NMR for strongly dipole-coupled nuclear spins to carry out controlled phase shifts for quantum-processing of the signal information.

In spite of a number of theoretical publications devoted to the Berry phase in NQR and NMR, experimental works are unavailable and the experiments in which the Berry phase would be recorded for pure NQR are not known except for the Tycko's one (monocrystal, $\eta=0$) and our experiment (powder, $\eta \neq 0$) [12].

In our publications [12,13] the Tycko's experiments for pure NQR were extended for the nuclei with spins $I=3/2$ and 1 for the substances in powder whose asymmetry parameter was not equal to zero. These our works were the first attempts at practical use of the Berry phase manifestation in pure NQR to obtain the asymmetry parameter of the EFG tensor on the nuclei with spin $I=3/2$ in powdered substances.

* Corresponding author.

E-mail addresses: glotova@amu.edu.pl (O. Glotova), natalya39@list.ru (N. Ponamareva), sinyavsky_physics@bga.gazinter.net (N. Sinyavsky), nogaj@amu.edu.pl (B. Nogaj).

Lisin et al. [14] investigated by NMR the geometric phase of a system with two levels, whose cyclic evolution was stimulated by means of “off-resonance” 2π -pulse. These authors proposed observations of the influence of the Berry phase without macroscopic rotation of a sample, for example, making use of the fact that the adiabatic evolution of a spin system can be achieved by rotation of nuclear magnetization in spin space induced by r.f. pulse sequence application. Such a rotation is the nutation of magnetization around the effective field. The frequency of nutation and positions of the nutation axis depend on the r.f. amplitudes of excitation pulses and on the frequency of detuning from the resonance. The Berry phase difference in different states of the Hamiltonian in a rotating frame system of reference results in the phase shift dependent on the orientation and asymmetry of FEG tensor, which is manifested in the experiment.

The aim of the present work was to check the effect of the open-ended trajectory of motion of nuclear magnetization on the phase of NQR signals of the sample in the off-resonance pulse NQR experiments carried out on the nuclei of spins $I=1$ and $3/2$ in powdered samples.

2. Theory

The trajectory of motion of nuclear magnetization in the spin space induced by the r.f. pulse is defined by nutation of magnetization around the effective field, depending on the amplitude of the excitation pulse and the frequency detuning from the resonance (Fig. 1).

Let's consider the nuclei of spin $I=1$ in an electric field with non-axial tensor symmetry of the electric field gradient $\eta \neq 0$. The energy levels in this case are not degenerated. To assess the influence of r.f. pulse with frequency detuning $\Delta\omega$ from the resonance on the spin system it is necessary to solve the non-stationary Schrödinger equation for the time interval $0 \leq t \leq t_w$, where t_w is the pulse duration. The wave function can be presented as [15]

$$\Psi = \sum_{m=-1}^{m=+1} C_m(t) \varphi_m \exp(-iE_m t/\hbar) \quad (1)$$

where φ_m are the orthogonal eigenfunction of quadrupole Hamiltonian H_Q , and E_m is the energy of quadrupole interaction energy level.

For the spin $I=1$ and for the asymmetry parameter $\eta \neq 0$, the frequencies of transitions between the three energy levels can be expressed as $\nu_{\pm} = eQq_{xx}/4(3 \pm \eta)$. The coefficients $C_m(t)$ in expression (1) are obtained by solving the non-stationary Schrödinger equation:

$$i\hbar \dot{\psi} = [H_Q + H_1(t)]\psi \quad (2)$$

in the interval $0 \leq t \leq t_w$. Using the property of orthogonality of the eigenfunctions φ_m ($m=+1, -1, 0$) and making use of (2), we

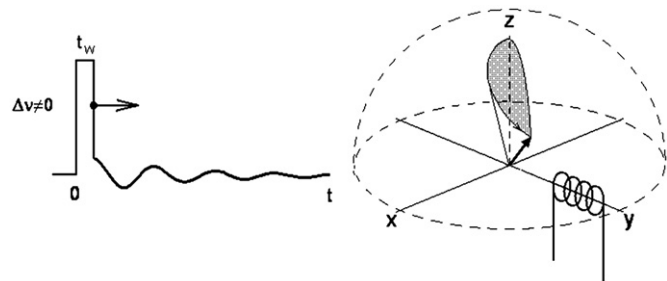


Fig. 1. Off-resonance r.f. pulse of variable duration and movement trajectory of nuclear magnetization of the isochromatrical group.

get the equation system for C_m :

$$i\hbar \frac{dC_m}{dt} = \sum_n V_{mn} C_n e^{-i\omega_{mn}t} \quad (3)$$

where $\omega_{mn} = E_m - E_n/\hbar$ are the frequencies of transitions between the energy levels. The Hamiltonian of interaction with r.f. field is defined as $\bar{H}_1 = -\gamma\hbar(\vec{B}_1 \cdot \vec{I})\cos\omega t$, where $\omega = \omega_{\pm} + \Delta\omega$ —the spectrometer frequency.

The matrix H_1 in H_Q —representation has the form: $H_1 = -\gamma\hbar B_1 \cos\omega_0 \cdot t V_{kn}$, where V_{kn} is the matrix with elements:

$$V_{kn} = \begin{pmatrix} 0 & \cos\theta & \sin\theta\cos\varphi \\ \cos\theta & 0 & -i\sin\theta\sin\varphi \\ \sin\theta\cos\varphi & i\sin\theta\sin\varphi & 0 \end{pmatrix} \quad (4)$$

Let's consider the excitation of a signal at the frequency ω_{+} . Then in the rotating frame the Hamiltonian of interaction with a r.f. field is: $H_1 = -\gamma\hbar B_1 \cos\omega t V_{mn} e^{i\omega_{mn}t}$. When only the terms slowly varying in time are left, we obtain:

$$H_1 = \begin{pmatrix} 0 & 0 & A_x e^{i\Delta\omega t} \\ 0 & 0 & 0 \\ A_x e^{-i\Delta\omega t} & 0 & 0 \end{pmatrix} \quad (5)$$

where $\Delta\omega$ is the spectrometer frequency detuning from the resonance, $A_x = \gamma B_1/2 \sin\theta \cos\varphi$, $A_y = \gamma B_1/2 \sin\theta \sin\varphi$ and $A_z = \gamma B_1/2 \cos\theta$.

The solution for $C_m(t)$ can be expressed in terms of the initial coefficients $C_m(0)$ and matrix R as follows: $C(t) = RC(0)$, where $C(0) = [C_{+1}(0) C_{-1}(0) C_0(0)]$ and

$$R = \begin{bmatrix} (\cos \frac{\alpha}{2} t - i \frac{\Delta\omega}{\alpha} \sin \frac{\alpha}{2} t) e^{i\Delta\omega t/2} & 0 & i \frac{2A_x}{\alpha} \sin \frac{\alpha}{2} t e^{i\Delta\omega t/2} \\ 0 & 1 & 0 \\ i \frac{2A_x}{\alpha} \sin \frac{\alpha}{2} t e^{-i\Delta\omega t/2} & 0 & (\cos \frac{\alpha}{2} t + i \frac{\Delta\omega}{\alpha} \sin \frac{\alpha}{2} t) e^{-i\Delta\omega t/2} \end{bmatrix} \quad (6)$$

Here $\omega_n = \sqrt{\Delta\omega^2 + 4A_x^2} = \sqrt{\Delta\omega^2 + \omega_1^2}$ is the nutation frequency; $\omega_1 = \gamma B_1 \sin\theta \cos\varphi$ the Rabi frequency.

The total phase of the induction signal consisting of geometric and dynamic phases can be calculated by means of the formula from [16]:

$$\Phi_t = \Phi_g + \Phi_d = \arg \langle \psi(0) | \psi(t_w) \rangle \quad (7)$$

where the non-cyclic geometric phase is

$$\Phi_g = \arctan \left[\frac{\Delta\omega}{\omega_n} \tan \left(\frac{\omega_n t_w}{2} \right) \right] - \frac{\Delta\omega t_w}{2} = \frac{1}{2} \Omega \quad (8)$$

and $\Phi_d = \arctan[\Delta\omega/\omega_n \tan(\omega_n t_w/2)]$ is the total phase, $\Phi_d \Delta\omega t_w/2$ the dynamic phase and Ω the solid angle made by the trajectory of the end of the vector of magnetization on the sphere and the geodetic curve passing through the final and initial points (Fig. 1). The value of this solid angle Ω can be calculated also only on the basis of geometry using the integral:

$$\Omega = 2 \int_{\theta_1}^{\theta_2} \sin\theta \arccos \left(\frac{\tan\theta_1}{\tan\theta} \right) d\theta \quad (9)$$

where $\theta_1 = \arctan[\cos(\omega_n t_w/2) \tan\theta_2]$ and $\theta_2 = \arccos(\Delta\omega/\omega_n)$.

Integration of expression (9) results in the formula for the solid angle:

$$\Omega = 2 \arctan \left[\frac{\Delta\omega}{\omega_n} \tan \left(\frac{\omega_n t_w}{2} \right) \right] - \Delta\omega t_w \quad (10)$$

On condition that the start of the recording is synchronized with the end of the r.f. pulse, the induction signal induced in the receiver coil set at ω_{+} ($I=1$) turns out [17] to be proportional to

Download English Version:

<https://daneshyari.com/en/article/5420664>

Download Persian Version:

<https://daneshyari.com/article/5420664>

[Daneshyari.com](https://daneshyari.com)