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RELAXATION OF ^{53}Cr SPIN ECHO SIGNALS IN $Cd_{0.985}Ag_{0.015}Cr_2Se_4$

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The frequency dependences of the relaxation times of NMR spin echo signals of quadrupole nuclei ^{53}Cr at $t_e = \tau$ and $t_e = 3\tau$ in ferromagnetic semiconductor $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ were investigated at temperature 4.2 K. It was shown that there are two kinds of the quadrupole nuclei ^{53}Cr, which have quite different relaxation times. The existence of two kinds of the nuclei ^{53}Cr was connected with doping of the cadmium selenochromite with Ag^+ ions.

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1. Introduction

The pulse-NMR method is one of the powerful techniques for the study of the spin dynamics in magnetically ordered materials. In the case of the quadrupole nuclei with spin $I = 3/2$ (for example ^{53}Cr nuclei), when the quadrupole interactions of the nuclei do not equal zero, the two echo signals may be observed at $t_e = \tau$ and $t_e = 3\tau$ [1–3]. The first echo signal V_τ is the usual Hahn echo and the NMR spectrum $V_\tau(\nu)$ recorded with the aid of this echo reflects all NMR spectral lines of the quadrupole nuclei. However, the spectrum NMR $V_{3\tau}(\nu)$ recorded with the aid of the echo at $t_e = 3\tau$ consists of the NMR resonance frequencies, whose values are determined by the hyperfine magnetic interaction only [1–3]. The aim of this paper is to analyze the relaxation of the $V_\tau(\nu)$ and $V_{3\tau}(\nu)$ echo signals of the quadrupole nuclei ^{53}Cr in ferromagnetic semiconductors $Cd_{0.985}Ag_{0.015}Cr_2Se_4$.

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2. Experimental results and discussion

The NMR measurements were made on the polycrystalline multidomain sample $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ at $T = 4.2$ K in zero static external magnetic field. The experimental results are shown in Fig. 1 and Fig. 2. The analysis of the obtained experimental results were provided assuming that the time fluctuations in the electron magnetization $\boldsymbol{M}_e$ due to the fluctuations in the hyperfine magnetic and quadrupole interactions lead to the relaxation of the spin echo signals. Assuming the frequency τ_{ce}^{-1} of the time fluctuations of $\boldsymbol{M}_e$ is smaller than NMR resonance frequencies ν_i and so remaining in the fluctuating hyperfine magnetic and electric quadrupole Hamiltonians the secular terms only we obtained the following expression for the relaxation rate of the echo signal $V_\tau(\nu)$:

$$T_2^{-1}(\tau, \nu_i) = A + B_i \sin^2(2\theta_0), \tag{1}$$

where $i = 1, 2, 3$; ν_1 is the resonance frequency of the NMR transition $\pm 1/2 \longleftrightarrow \mp 1/2$; $\nu_{2,3}$ are the resonance frequencies of the NMR transitions $\pm 3/2 \longleftrightarrow \pm 1/2$. In Eq. (1) the angle θ_0 is the angle between the local trigonal axis and a direction of the electron magnetization vector $\boldsymbol{M}_e$. The NMR resonance frequencies ν_i are uniquely determined by the angle θ_0. The solid lines shown in Fig. 1 represent the theoretical frequency dependences obtained from the best fit of Eq. (1) to the observed values of $T_2(\tau, \nu)$. As is seen, the theoretical curves agree well with the experimental results. In the secular approximation we obtained the following expression for the relaxation of the multiquantum echo signal $V_{3\tau}(\nu_1)$:

$$T_2^{-1}(3\tau, \nu_1) = 3T_2^{-1}(\tau, \nu_1). \tag{2}$$

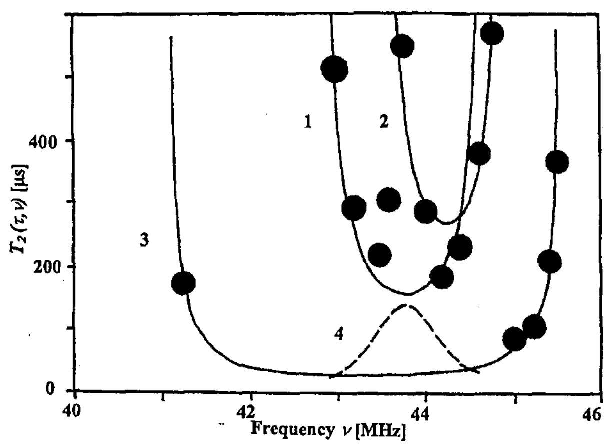

Fig. 1. Frequency dependence of the relaxation time $T_2(\tau, \nu)$ of the ^{53}Cr nuclei in $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ at $T = 4.2$ K. The solid lines are the theoretical curves obtained from the best fit of Eq. (1) to the measured values of $T_2(\tau, \nu)$. Curve 1 is the dependence $T_2(\tau, \nu_1)$; curves 2 and 3 are the dependences $T_2(\tau, \nu_2)$ and $T_2(\tau, \nu_3)$. Broken line 4 is the theoretical curve $T_2(\tau, \nu_1)$ obtained in the nonsecular approximation.

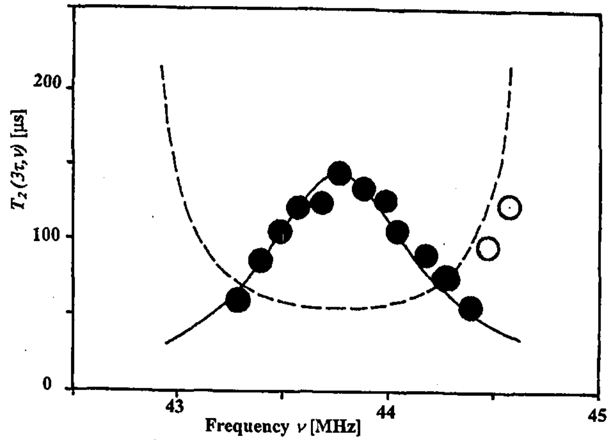

Fig. 2. Frequency dependence of the relaxation time $T_2(3\tau, \nu)$ of the ^{53}Cr nuclei in $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ at $T = 4.2$ K. The solid lines are the theoretical curves obtained from the best fit of Eq. (3) to the measured values of $T_2(3\tau, \nu)$ (black circles). The broken line is the theoretical curve $\frac{1}{3}T_2(\tau, \nu_1)$ obtained with the parameters defined from curve 1 in Fig. 1.

As is seen from Fig. 2 only two experimental points (the open circles) coincide with frequency dependence (2). In order to understand the source of this discrepancy we considered the relaxation of the echo signal at $t_e = 3\tau$ assuming that the time fluctuations of M_e are not small ($\tau_{ce}^{-1} \gg \nu_i$) and retaining in the fluctuating Hamiltonians the nonsecular terms too. The obtained expression for the relaxation rate of spin echo $V_{3\tau}(\nu_1)$ has the form

$$T_2^{-1}(3\tau, \nu_1) = C + D\cos^2(2\theta_0). \tag{3}$$

As is seen from Fig. 2 the dependence (3) well describes the observed dependence of $T_2(3\tau, \nu_1)$.

The obtained results suggest that in $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ there are two kinds of the ^{53}Cr nuclei which have quite different relaxation channels ("secular" and "nonsecular"). The nuclei of the first kind ^{53}Cr(I) give the main contribution to the echo signal $V_\tau(\nu)$. In the echo signal $V_{3\tau}(\nu)$ these nuclei are observed only at $\nu > 44.5$ MHz (the open circles in Fig. 2). The nuclei of the second kind ^{53}Cr(II) have very short relaxation time $T_2(\tau, \nu)$ and so they do not give the contribution to the observed echo signal $V_\nu(\tau)$. We may observe these nuclei only with help of the echo $V_{3\tau}(\nu)$ (the black circles in Fig. 2). In order to understand why the nuclei ^{53}Cr(II) do not give the contribution to the echo signal at $t_e = \tau$ we considered the nonsecular relaxation of the echo signal $V_\tau(\nu_1)$. The broken line in Fig. 1 represents the obtained theoretical curve. As is seen the relaxation of the echo signal $V_\tau(\nu)$ from the nuclei ^{53}Cr(II) is indeed smaller one for the nuclei ^{53}Cr(I). It is reasonable to assume that the existence of two kinds of the nuclei ^{53}Cr in $Cd_{0.985}Ag_{0.015}Cr_2Se_4$ is connected with the doping of $CdCr_2Se_4$ with Ag^+ ions. The doping of the cadmium selenochromite with silver ions produces, as a result of electric charge compensation, the Cr^{4+} impurities. The different relaxation channels for the nuclei

^{53}Cr(I) and ^{53}Cr(II) are probably connected with the dynamical nature of the Cr^{4+} defects. We assume that electron exchange between the Cr^{4+} and Cr^{3+} ions sited inside of the defect region leads to the rapid fluctuations in the local electron magnetization ($\tau_{ce}^{-1} \gg \nu_i$) so the nuclei ^{53} Cr(II) located in these defect regions "feel" due to the hyperfine and quadrupole interactions the rapidly fluctuating electron magnetization. The rate of local fluctuations of $\boldsymbol{M}_e$ for the nuclei ^{53}Cr(I) which are sited far from defects is small, then ν_i and the relaxation of the echo signals from these nuclei is "secular".

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