Spin-lattice relaxation in $Bi_4Ge_3O_{12}$ - quadrupole and magnetic mechanisms

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Abstract

The nuclear spin-lattice relaxation was studied in the $Bi_4Ge_3O_{12}$ single crystal by ^{209}Bi NQR. Experimental recovery curves for all ^{209}Bi NQR transitions of nuclear spin I=9/2 in the temperature range 10-230K were described by the single effective spin-lattice relaxation time $T_1{}^*$. The temperature dependence of $1/T_1{}^*$ was close to T^n law with n=2.4 - 2.7. Theoretical treatment was given for the nuclear spin-lattice relaxation in the multi-level system for the case of single-axial crystalline electric field. Both quadrupole and magnetic mechanisms of relaxation were taken into account. It was shown that magnetic mechanism contributes noticeably to the spin-lattice relaxation in $Bi_4Ge_3O_{12}$ at T≤50K.

Key words: spin-lattice relaxation; ^{209}Bi NQR

1. Introduction

$Bi_4Ge_3O_{12}$ belongs to the class of complex Bi-oxides (α - Bi_2O_3, Bi_3O_4Br) which exhibits unusual magnetic properties [1]. Local ordered magnetic fields of about 30 G were revealed in the $Bi_4Ge_3O_{12}$ single crystal by ^{209}Bi NQR experiments [2]. In the present paper the nuclear spin-lattice relaxation was studied in the $Bi_4Ge_3O_{12}$ single crystal by ^{209}Bi NQR spin-echo technique.

The spin-lattice relaxation rate was measured using the saturation recovery method. The recovery curves for all four transitions are successfully fitted by single exponential function with characteristic time $T_{1i}{}^*$. The temperature dependencies of $1/T_{1i}{}^*$ are close to T^n law with n = 2.4-2.7.

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2. Results and discussion

The theoretical study of the relaxation process is based on the master equation:

$$\frac{dN_i}{dt} = \sum_j (W_{ij} N_j - W_{ji} N_i). \qquad (1)$$

where N_i is the population of the i-th level, W_{ij} is the probability of transition from the j-th to the i-th level, the sum over j is taken for all allowed transitions (△ m =± 1 and △ m =± 2) between the levels for the spin I=9/2 . The probabilities for these transitions were calculated using the formulae [3]:

$$\frac{W^m_{m\pm1}}{W_1} = (2m \pm 1)^2 (I \mp m) K \qquad (2)$$

$$\frac{W^m_{m\pm2}}{W_2} = (I \mp m)(I \mp m - 1)(I \pm m + 2) K \qquad (3)$$

where

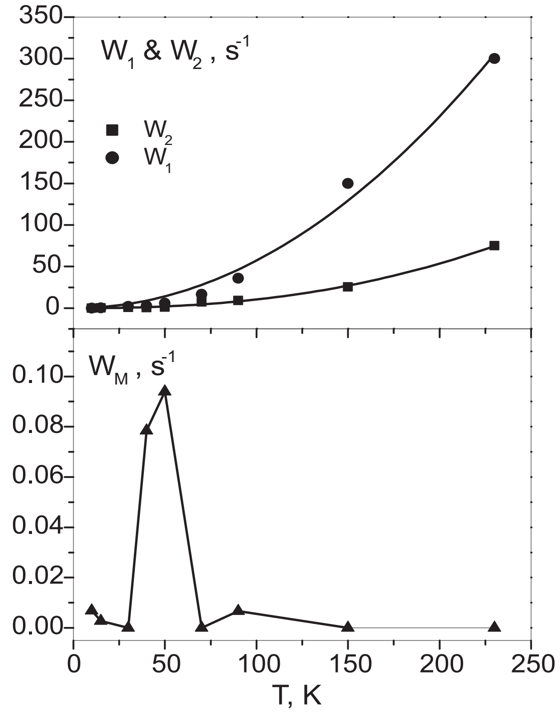

Fig. 1. The temperature dependence of parameters W_1, W_2 (top) and W_M (bottom). Solid lines are the best fit by the power law: W(T) T^n with n = 2.0(1) for W_1 and n = 2.4(1) for W_2. In W_M(T) plot solid line is a guide for the eye.

$$K = \frac{(I \pm m + 1)}{2I^2(2I-1)^2}, \tag{4}$$

W_1 and W_2 are unknown quantities.

Transition probabilities for the $\triangle$ m =$\pm$ 1 magnetic relaxation processes were defined in [4]:

$$\frac{W^m_{m\pm 1,M}}{W_M} = (I \mp m)(I \pm m + 1). \tag{5}$$

The relaxation process can be written in the following form:

$$\frac{n_i}{n_0} = \alpha_i + \sum_{j=1}^{4} c_j a_{ij} e^{-\lambda_j \tau} \tag{6}$$

the values of λ_j determine the time scale of relaxation, the pre-exponential factors $c_j a_{ij}$ are responsible for the form of the relaxation curve. An effective calculated relaxation time $T_1{}^i$ for the i-th transition is defined by:

$$\frac{d}{dt}\frac{n_i}{\alpha_i n_0}_{t=0} = -\frac{W_i}{108\alpha_i}\sum_{j=1}^{4} a_{ij} c_j \lambda_j \equiv \frac{1}{T_1^{(i)}}. \tag{7}$$

The fitting procedure for the effective relaxation times $T_1{}^{(i)}$ and $T_{1i}{}^*$ was carried out numerically to define a temperature dependence of parameters W_1,W_2 and W_M. The details of theoretical calculation are presented in [5].

The effective calculated relaxation rates $1/T_1{}^{(i)}$ clarify the meaning of the effective experimental relaxation rates $R_{1i}{}^*$ for the multi-level system as the weighted sum of the "true" relaxation rates. The 'dominant' contribution (quadrupole or magnetic) determines the relaxation process, therefore the time evolution of the multi-level system to equilibrium state can be described by a single time constant.

As follows from Fig.1, the parameters W_1 and W_2 grow rapidly close to the parabolic law similar to the temperature behavior of the experimental effective relaxation rates $R_{1i}{}^*$. It is necessary to note that throughout the studied temperature range 10 - 230 K the values of W_2 parameter were noticeably smaller than those of W_1. So the magnitude of the ratio $\gamma = W_2/W_1$ never exceeded 0.5. In some previous works γ was taken equal to 1 [6], [7]. The bottom panel in Fig. 1 shows the temperature dependence of W_M. Basing on the obtained results we can conclude that at T $\geq$ 70 K the spin-lattice relaxation in $Bi_4Ge_3O_{12}$ is governed mainly by the quadrupolar mechanism. At T $\leq$ 50 K the contribution of the magnetic mechanism is quite noticeable. According to (5), the probabilities of transitions governed by the magnetic mechanism attain rather large. At low temperatures, when W_1 is not large (Fig.1), even small values of W_M contribute significantly to the relaxation.

3. Conclusion

It was demonstrated that for the case of single-axial EFG and weak local magnetic field realized in $Bi_4Ge_3O_{12}$ it is possible to separate the contributions of quadrupole and magnetic mechanisms to relaxation. At T $>$ 70 K the spin-lattice relaxation is governed mainly by quadrupole mechanism, while at low temperatures the contribution of magnetic mechanism is noticeable.

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