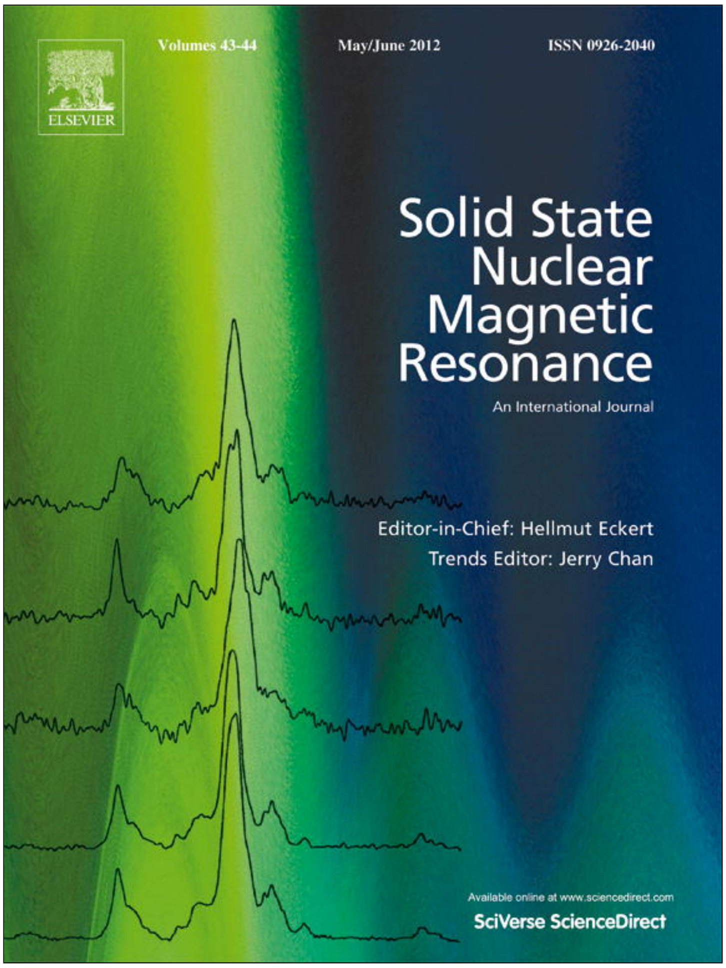
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Hahn-echo decay for exchange-coupled nuclear spins in solids

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ABSTRACT

In this paper we present a simple model to calculate the Hahn-echo decay of the exchange-coupled nuclear spins in solids. Satisfactory agreement between the calculated and experimentally observed echo decay of the exchange-coupled spins of ^{203}Tl and ^{205}Tl isotopes in thallium chloride TlCl and thallium tantalum sulfide $TlTaS_3$ is obtained.

1. Introduction

The spin echo technique is one of the most powerful methods for the investigations of quantum dynamics of the two- and multi-level systems. It has recently attracted special attention in studying the spin fluctuations in the cuprate superconductors [1–3], nanoelectronics [4,5], multi-quantum effects in ferromagnetics [6], long-lived echoes in magnetically dilute systems [7] and particularly in quantum information processing [8–11]. Among the quantum computer concepts under discussion, nuclear spins are considered to be the ideal quantum bits (qubits) embodied in crystal lattices. The spin echo technique is the main tool to measure the spin decoherence time, a crucial parameter in spintronics and quantum computing where a nuclear spin is used as a qubit.

In this paper we analyze the decay of Hahn echoes of ^{203}Tl and ^{205}Tl in solid thallium compounds, in which the NMR line shape is known to be dominated by strong indirect exchange interactions among nuclear spins [12–23] described by the Hamiltonian [24–27]

$$H_{ex}=\sum_{k<l}J_{kl}I_kI_l=\sum_{k<l}J_{kl}[I_k^zI_l^z+\frac{1}{2}(I_k^+I_l^-+I_k^-I_l^+)] \quad (1)$$

where Eq. (1) represents the scalar term of the exchange Hamiltonian, and J_{ik} are the exchange coupling constants. Tensor terms of the exchange Hamiltonian were shown to be much smaller compared with the scalar term [12] and thus neglected. The exchange interactions are realized across the overlapping electron clouds of atoms [25]. Van Vleck has shown [27] that like and unlike spins yield different contribution to the second moment of the NMR line,

$$M_2=\int_{-\infty}^{\infty}(\nu-\nu_0)^2f(\nu)d\nu\Big/\int_{-\infty}^{\infty}f(\nu)d\nu=\mathrm{Tr}\left[H_{ex}/h\sum_i I_{ix}\right]^2\Big/\mathrm{Tr}\left(\sum_i I_{ix}\right)^2, \quad (2)$$

where ν_0 is the first moment of the NMR line shape $f(\nu)$. For two identical nuclei, the exchange term $J_{12}\vec{I}_1\vec{I}_2$ commutes with $I_{1x}+I_{2x}$, and therefore exchange interaction among like spins does not contribute to the second moment M_2 but makes a contribution to the fourth moment M_4 of the NMR line. Since $M_4\geq 3M_2^2$, it results in the exchange line narrowing, the well known effect in the solid state NMR [24–26]. In such a case the line shape is predicted to be Lorentzian and to have long wings, which are often not observed experimentally, being lost in the noise. The measured second moment in strong exchange-coupled systems may be reduced (though theoretically [24,25] is should not be changed). This fact restricts the application of the method of moments for NMR line shape analysis in exchange-coupled systems. This situation is similar to that of invariance of the second moment in solids with molecular dynamics [25], though formally the second moment of the NMR spectrum is not dependent on frequency or character of mobility of magnetic nuclei [25], but actually their mobility results in the NMR spectrum narrowing and in reduction of the experimentally measured M_2 [25].

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If the two nuclei are not identical, one can approximate the exchange coupling as $J_{12}I_{1z}I_{2z}$, which does not commute with I_{2z} and I_{1z} and therefore increases the second moment [24,25,27], resulting in line broadening. The exchange narrowing of the NMR line is proportional to the natural abundance of the like isotope, while the exchange broadening of the NMR line is proportional to the abundance of the unlike isotope. For thallium, the natural abundances are $f=29.5\%$ for ^{203}Tl and $(1-f)=70.5\%$ for ^{205}Tl with $(1-f)/f=2.39$, which makes the aforementioned effects readily observable. The first experimental evidence of such effects (in Tl_2O_3) was reported in the classic paper of Bloembergen and Rowland [12]; then it was observed by the other authors (e.g., [13–23]). The contribution of the dipole–dipole interactions between nuclear spins to the line shape is by one to two orders of magnitude smaller than that of the exchange coupling and therefore may be neglected [12–23].

It is known that the Hahn echo after the pulse sequence $(\pi/2)_0-\tau-(\pi)_{90}-\tau'$-echo is not formed in the presence of homonuclear exchange interactions, whose time evolution cannot be reversed by the Hahn-echo sequence. However, the heteronuclear interactions $J_{12}I_{1z}I_{2z}$ are effectively refocused by the Hahn-echo sequence. The Hahn-echo decay is caused by the interactions that are quadratic in the nuclear spin operators of the resonant nuclei, such as dipolar and homonuclear indirect exchange couplings, since the sign of these interactions is not changed by the second π pulse.

To calculate the Hahn-echo decay, the explicit information about the aforementioned interactions is needed. The effect of the indirect exchange interaction $A\vec{I}_i\vec{I}_j$ among the resonant and off-resonant nuclei on the NMR line shape has been discussed both theoretically and experimentally in a number of papers [12,13,26]. Such an effect has traditionally been considered in terms of the method of moments [25,26]. However, the aforementioned difficulties in application of the moment method for analysis of the exchanged-coupled spin systems in solids urge one to turn to the NMR line shape analysis instead of analysis of its moments. On the other hand, the line shape calculation is usually a multi-particle problem that does not have an exact solution. Due to this fact one has to discuss some approximate theoretical models, the reliability of which is probed by the comparison with the experimental data. In this paper we present a simple model that allows a satisfactory description of the experimentally observed spin echo decay of the exchange-coupled nuclear spins of ^{203}Tl and ^{205}Tl isotopes in thallium chloride TlCl and thallium tantalum sulfide $TlTaS_3$. Our model is based on the mechanism of exchange averaging of local magnetic fields on the resonance nuclei. [25,26] We suggest that the exchange flip–flop interactions $I^A_\pm I^A_\mp$ and $I^B_\pm I^B_\mp$ between resonant A and off-resonant B spins cause stochastic fluctuations of z-components of A and B spins that result in averaging of the interaction, which are caused by the term $I^A_zI^B_z$ in the exchange interaction Hamiltonian. The degree of such averaging depends on the exchange coupling between like resonant and like non-resonant spins; the averaging results in the NMR line narrowing. We suggest that this mechanism is responsible for the observed Hahn-echo decay in exchange-coupled systems.

2. Experimental details

We studied Hahn-echo decay in two powder samples: thallium chloride TlCl and thallium tantalum sulfide $TlTaS_3$. All ^{203}Tl and ^{205}Tl NMR measurements were carried out at room temperature using a Tecmag APOLLO pulse solid state NMR spectrometer, a Varian electromagnet and an Oxford superconducting magnet. The ^{203}Tl and ^{205}Tl NMR spectra and spin echo decays in TlCl were measured (i) at a resonance frequency of 28.7 MHz in magnetic fields 1.143 and 1.132 T for ^{203}Tl and ^{205}Tl isotopes, respectively, and (ii) at resonance frequencies 195.09 and 197.01 MHz in a magnetic field of 8.0196 T for ^{203}Tl and ^{205}Tl isotopes, respectively. The ^{203}Tl and ^{205}Tl NMR spectra and spin echo decays in $TlTaS_3$ were measured in magnetic fields of 1.143 and 1.132 T for ^{203}Tl and ^{205}Tl isotopes, respectively (at a resonance frequency of 28.7 MHz).

3. Results and discussion

^{203}Tl and ^{205}Tl spectra of TlCl are shown in Fig. 1, representing Lorentzian-like curves with full line widths at half maximum $\Delta\nu(^{205}\text{Tl})=6.1\pm0.1$ kHz and $\Delta\nu(^{203}\text{Tl})=8.8\pm0.1$ kHz and second moments $M_2(^{205}\text{Tl})=22.2\pm1.1$ kHz^2 and $M_2(^{203}\text{Tl})=45.8\pm2.1$ kHz^2. The latter were determined using a computer program for calculation of the second moments of the experimental spectra. Crystal structure of TlCl belongs to the cubic CsCl type (space group $Pm3m$) with Tl atoms in the corners of the cube and Cl atoms in its center. Each Tl has 6 nearest Tl neighbors at 3.85 Å, 12 next Tl neighbors at 5.44 Å, and 8 Cl neighbors at 3.33 Å. Owing to the cubic structure, thallium NMR spectra in TlCl does not show chemical shielding anisotropy. Since both thallium ^{203}Tl and ^{205}Tl isotopes have spin $I=1/2$, the quadrupolar contribution is absent. The contributions of the dipole–dipole coupling of nuclear spins to the second moment of thallium resonances, calculated using Van Vleck formula [27], are 1.21 kHz^2 for ^{203}Tl and 1.69 kHz^2 for ^{205}Tl, respectively, and are much smaller than the experimentally measured values. Thus one can conclude that the experimental second moments are predominantly determined by the indirect exchange coupling.

^{203}Tl and ^{205}Tl spectra of $TlTaS_3$ were published elsewhere [18,20]. They show $M_2=85$ and 100 kHz^2 for ^{205}Tl and ^{203}Tl isotopes, respectively in a magnetic field of 1.2 T. These values are much larger than those resulting from the contributions of the dipole–dipole interactions of nuclear spins ($\sim$1.2 kHz^2). The crystal structure of $TlTaS_3$ [18,20,28] is orthorhombic (space group $Pnma$). Thallium atoms occupy only one crystallographic position. Tl coordination by the S atoms is asymmetric, resulting in the significant chemical shielding anisotropy that dominates the spectra in high magnetic field [18,20]. Therefore in this paper we analyze the ^{203}Tl and ^{205}Tl NMR spectra and spin echo decays

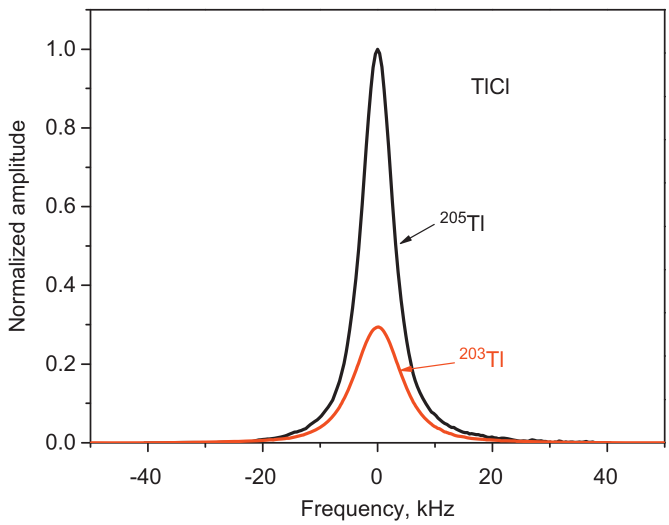

Fig. 1. ^{203}Tl and ^{205}Tl NMR spectra of TlCl in magnetic field of 8.0196 T.

in $TlTaS_3$ that were measured only at the low magnetic field, in which the contribution of the chemical shielding anisotropy to the line shape and relaxation parameters is negligible.

The experimental echo decays and their parameters are shown in Figs. 2–4 and in Table 1. Our system comprises unlike spins of two interacting ^{203}Tl and ^{205}Tl isotopes. For the sake of simplicity, we consider only scalar (isotropic) exchange terms, neglecting the anisotropic (pseudo-dipolar) indirect interaction and dipolar coupling that are usually much smaller than the scalar term [12–23].

The scalar term of spin Hamiltonian for the system consisting of two types of nuclei A and B is

$$H_{ex} = H^{AA} + H^{BB} + H^{AB} = \sum_{k,l} J_{kl} \vec{I}_k^A \vec{I}_l^A + \sum_{i,k} J_{kl} \vec{I}_k^B \vec{I}_l^B + \sum_{i,k} J_{kl} I_{zk}^A I_{zl}^B, \quad (3)$$

where J_{kl} are the exchange coupling constants and indexes A and B denote the resonant and off-resonant nuclei, respectively.

As it is mentioned above, the multi-particle character of the interaction Hamiltonian (3) does not allow one to calculate explicitly the spin echo decay. Therefore, discussing the echo decay, we model our system by a simplified nuclear spin Hamiltonian, which, in our opinion, reflects the main physical features of the problem under discussion [26,29,30]

$$H_{ex} \cong \sum_i \omega_i(t) I_{zi}^A. \quad (4)$$

Here the fluctuations of the resonance frequency ω_i of nucleus A_i appear, owing to the (i) exchange coupling between the resonant nuclei A and off-resonant nuclei B, (ii) exchange coupling among the resonant nuclei of the A type in the $AAA\ldots A$ spin subsystem and (iii) exchange coupling among the off-resonant nuclei of the B type in the $BBB\ldots B$ spin subsystem. The latter seem to be the main source of fluctuations. Some fluctuations of ω_i can also be caused by interaction of resonant nuclei with paramagnetic defects [32] or by spin–lattice interactions [1,2].

Since the approximate Hamiltonian (4) is the sum of the one-particle Hamiltonians, further for simplification we will consider the echo signal of a single spin. The spin-echo signal is described by the following equation [30]

$$V(t) = \mathrm{Re}\left\langle \exp i\left[\int_0^\tau \omega(t')dt' - \int_\tau^t \omega(t')dt'\right]\right\rangle. \quad (5)$$

Evaluations for different stochastic processes were considered in a number of articles [29–31,33–38]. All neighboring spins contribute to the change in the orientation of a certain spin. Due to the large number of these contributions, one can assume that this stochastic process is Gaussian. It has been shown that for a stochastic Gaussian spectral diffusion process the decay of the echo signal at 2τ is described by the equation [30,31]

$$V(2\tau) = \exp\left\{-\left(\frac{\Delta_A}{\omega_{ex}}\right)^2 (2\tau\cdot\omega_{ex} - 3 + 4\cdot\exp(-\tau\cdot\omega_{ex}) - \exp(-2\tau\cdot\omega_{ex}))\right\}, \quad (6)$$

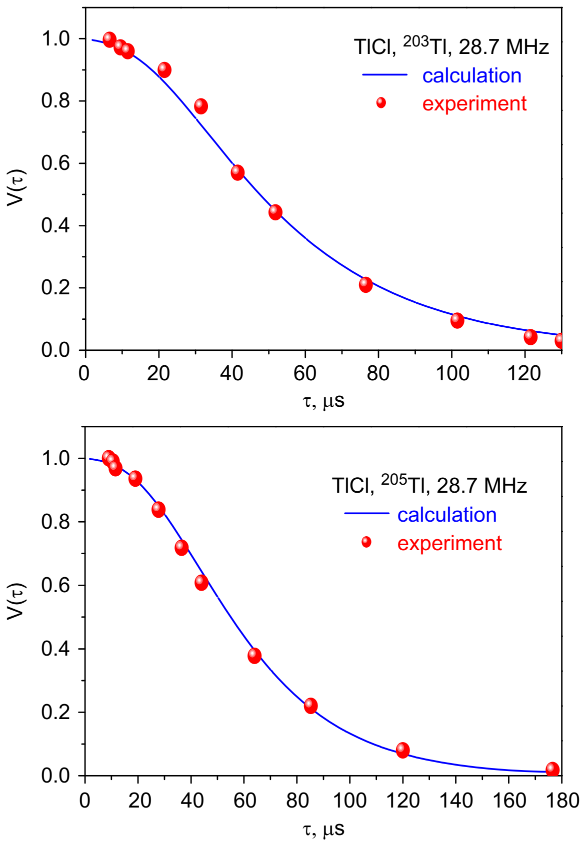

Fig. 2. The decay of Hahn-echo signals of ^{203}Tl (top) and ^{205}Tl (bottom) nuclei in polycrystalline TlCl at a resonance frequency $\nu = 28.7$ MHz. The circles are the experimental data, the solid line is the result of calculation using Eq. (6).

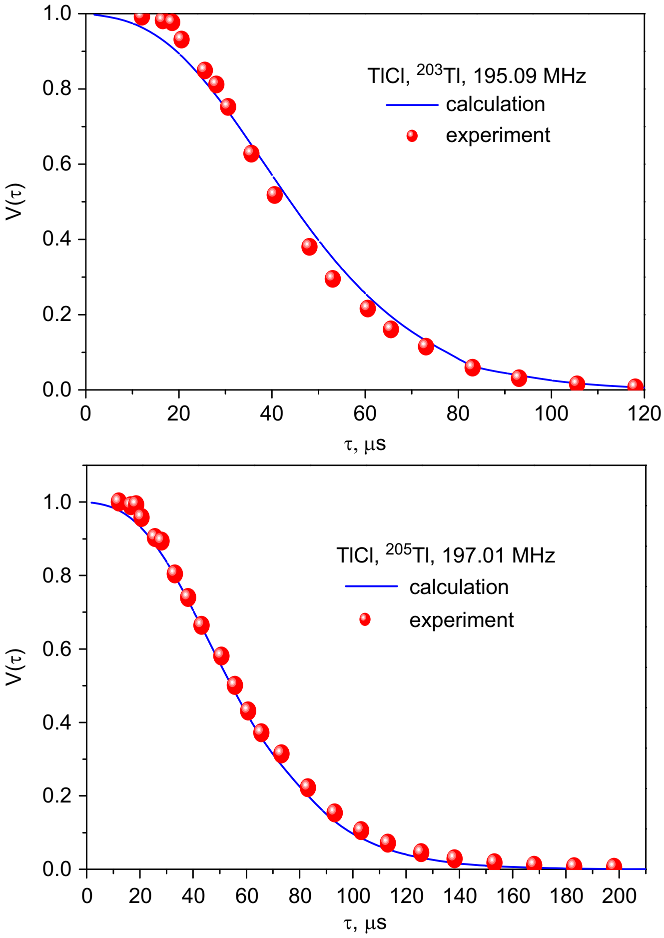

Fig. 3. The decay of Hahn-echo signals of ^{203}Tl (top) and ^{205}Tl (bottom) nuclei in polycrystalline TlCl at resonance frequencies $\nu = 195.09$ MHz for ^{203}Tl and $\nu = 197.01$ MHz for ^{205}Tl. The circles are the experimental data, the solid line is the result of calculation using Eq. (6).

where $\Delta_A = \langle \omega_i^2 \rangle^{1/2} = 2\pi\sqrt{M_{2A}}$. Here M_{2A} is the experimental second moment of the NMR line of the resonant nuclei A [30,31]. The parameter Δ_A describes the indirect exchange interaction between *A* and *B* spins. We note that, in our model, the frequency Δ_A is an independent fitting parameter.

In Eq. (6) ω_{ex} is the frequency of stochastic changes of the resonance frequency of nucleus A caused by the flip–flop jumps of both off-resonant and resonant spins. In [12,13,24,25] it was proposed to approximate the exchange frequency ω_{ex} as $\omega_{ex} \cong [(M_4 - 3M_2^2)/M_2]^{1/2} 2\pi$. In our model the frequency ω_{ex} is an independent fitting parameter.

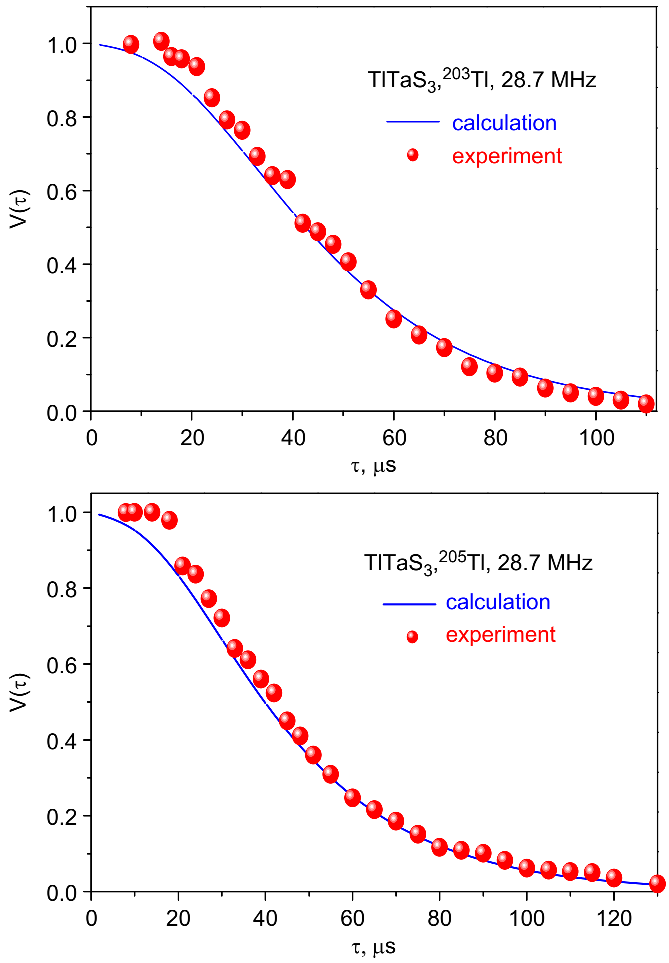

Fig. 4. The decay of Hahn-echo signals of ^{203}Tl (top) and ^{205}Tl (bottom) nuclei in polycrystalline $TlTaS_3$ at a resonance frequency $\nu = 28.7$ MHz. The circles are the experimental data, the solid line is the result of calculation using Eq. (6).

We used Eq. (6) for the fitting of the experimental Hahn-echo decays of ^{203}Tl and ^{205}Tl nuclei in polycrystalline TlCl and $TlTaS_3$ (Figs. 2–4). The values of Δ_A and ω_{ex}, obtained using Eq. (6), are given in Table 1. It should be noted that only these two parameters were used to fit the theory with the experimental data. In Table 1 we also present the spin–spin relaxation time T_2, which was determined as a point at which the echo amplitude falls to $1/e$ of its initial value.

One can find from the comparison of the theoretical and experimental curves (Figs. 2–4) that our simple model explains well the experimental data. From the obtained parameter Δ_A we find that $\Delta_A \approx 2\pi\sqrt{M_{2A}}$.

One can find from Table 1 that for TlCl $\omega_{ex}(^{203}\mathrm{Tl}) > \omega_{ex}(^{205}\mathrm{Tl})$ in both low and high magnetic fields. In our opinion, this finding may be explained as following: The main contribution to the frequency ω_{ex}, which determines the random fluctuations of the resonance frequency of nuclei A, is caused by the flip–flop processes of the off-resonant nuclei B. Since the natural abundance of ^{205}Tl isotope (70.5%) is larger than that of ^{203}Tl isotope (29.5%), the ^{205}Tl nuclei have more ^{205}Tl neighbors than ^{203}Tl neighbors. It will cause the flip–flop frequency of ^{205}Tl spins to be higher than that of ^{203}Tl spins, and, as a consequence, $\omega_{ex}(^{203}\mathrm{Tl}) > \omega_{ex}(^{205}\mathrm{Tl})$.

We notice that the value of ω_{ex} in TlCl increases with decreasing of the applied magnetic field, i.e., $\omega_{ex}(28.7\ \mathrm{MHz}) \geq \omega_{ex}(195.09\ \mathrm{MHz})$. Along with the above discussion, it means a suppression of the flip–flop processes of the off-resonant nuclei with increasing magnetic field. Such an effect may be caused by inhomogeneous broadening that increases with the increased magnetic field. Table 1 shows that this effect is stronger for the ^{205}Tl nuclei.

Differently from TlCl, $TlTaS_3$ shows $\omega_{ex}(^{205}\mathrm{Tl}) > \omega_{ex}(^{203}\mathrm{Tl})$. Such a difference may be caused by two reasons. First, $TlTaS_3$ reveals noticeable chemical shielding anisotropy [18,20,28] which, similar to the above-mentioned inhomogeneous broadening, results in a suppression of the flip–flop processes of the off-resonant nuclei. Second, $TlTaS_3$ reveals noticeable exchange coupling between ^{203,205}Tl and ^{181}Ta (natural abundance 99.98%) spins [18,20,28], which produces an additional local magnetic field on the off-resonant nuclei B; a spread of this field results in a suppression of the flip–flops among B nuclei.

Summarizing, we developed a simple model for the Hahn-echo decay of the exchange-coupled nuclear spins in solids. This model satisfactory describes the experimental data on ^{203}Tl and ^{205}Tl echo decay in thallium chloride TlCl and thallium tantalum sulfide $TlTaS_3$. The model will help in calculating the spin decoherence time in indirect exchange coupled nuclear systems that are considered for the use in spintronics and quantum computing.

Table 1
Parameters Δ_A, ω_{ex} (in units of angular frequency) and T_2, obtained from experimental data. Here M_{2A} is the experimental second moment of the NMR line of the resonant nuclei A.

Compound	Resonance frequency (MHz)	Resonant nucleus A	$2\pi\sqrt{M_{2A}}$ (rad × kHz)	ω_{ex}(rad × *kHz*)	Δ_A (rad × kHz)	T_2 (μs)
TlCl	28.7	^{203}Tl	25.15	50 (4)	25 (2)	59
TlCl	28.7	^{205}Tl	20.1	30 (3)	21 (2)	67
TlCl	195.09	^{203}Tl	42.5	35 (3)	42 (3)	52
TlCl	197.01	^{205}Tl	29.6	30 (3)	30 (3)	64
$TlTaS_3$	28.7	^{203}Tl	62.8	95 (6)	62 (5)	52
$TlTaS_3$	28.7	^{205}Tl	57.8	110 (7)	61 (5)	49

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