

Solid-Echo in Solids with Molecular Motions: Effects of Nonzero Pulse Widths

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Abstract. The effects of nonzero pulse widths on the solid-echo signals in solids with molecular motions have been investigated. It has been shown that in the slow-motion region ($M_2 \tau_c^2 \approx 1$) the amplitude of the echo signal is reduced and the maximum of the echo signal is shifted to the end of the second pulse. Comparison of the developed theory with experimental results obtained on polycrystalline C_6H_6 and NH_4Cl demonstrates good agreement between them.

1 Introduction

The “solid-echo” technique $90_y^\circ - \tau - 90_x^\circ - \text{Acq}(t)$, proposed by Powles, Mansfield and Strange [1, 2] provides a means of circumventing the receiver dead-time problem which would otherwise cause the omission of a major part of the free induction decay (FID) signal. This technique is a powerful method of studying molecular structure and dynamics in solid state by 1H nuclear magnetic resonance (NMR). General reviews of application of deuterium solid-echo technique have been recently published [3, 4].

It is now well-established that solid-echo signal can be significantly distorted in the so-called slow-motion region and the nature of this distortion may be used to obtain information about the motional mechanism and the motional parameters of nuclei in solids. At the present time, there have been published a great number of papers describing analysis and application of “solid-echo” technique to the molecular dynamics investigations in solids [4–14]. It has been assumed almost in all of these papers that radio-frequency (RF) pulses are the delta-functions. In the delta-function approximation of the RF pulses it is assumed that for the amplitude ω_1 and the widths t_i ($i = 1, 2$) of the RF pulses $\omega_1 t_i = \text{const}$ at $t_i \rightarrow 0$ and $\omega_1 \rightarrow \infty$. This approximation is not good enough for solids because the experimental RF pulses have values of their amplitudes comparable with the

NMR linewidth in solids. The effects of the finite RF pulses on the solid echoes with rigid lattice have been discussed in [15–17]. It has been shown that the internal interactions (dipolar or quadrupole) of the nuclei during the hard RF pulses produce the shift of the echo signal maximum from the time 2τ and the maximum of the echo signal is observed at $t_e = 2\tau + t_2 - t_1/2$ [15–17].

In this paper we consider the effects of the dipolar interactions of the spins during the RF pulses on the solid-echo signal for the case when there are the molecular motions in solids.

2 Theory

Let us consider an ensemble of nuclear spins in a high static magnetic field \mathbf{B}_0 ($\mathbf{B}_0 \parallel OZ$). The equilibrium density operator ρ at $t = 0$ can be written in high-temperature approximation as [18–20]:

$$\rho(0) = \beta I_z, \quad (1)$$

where $\beta = \hbar\omega_0/kT$, $\omega_0 = \gamma B_0$ is the Larmor frequency (γ is the magnetogyric ratio of nuclei) and T is the temperature of lattice.

After the first RF pulse (RF field in a coordinate frame rotating with the Larmor frequency about B_0 is B_{1j}) the density operator in the rotating coordinate frame is given at the time t_1 (t_1 is the width of the first RF pulse) by [18–20]

$$\rho(t_1) = \hat{U}_1(t_1, 0)\rho(0), \quad (2)$$

where the evolution superoperator $\hat{U}_1(t_1, 0)$ is given by

$$\hat{U}(t_1, 0) = 1 - i \int_0^{t_1} \hat{L}_1(t') dt' - \frac{1}{2} \int_0^{t_1} dt'' \int_0^{t''} \hat{L}_1(t'') L_1(t') dt' + \dots \quad (3)$$

and the Liouville superoperator $\hat{L}_1(t')$ has the following form:

$$\hat{L}_1(t') = [-\omega_1 I_Y + \mathcal{H}_Z(t'), \dots]. \quad (4)$$

Here $\omega_1 = \gamma B_1$ and $\mathcal{H}_Z(t')$ is the secular part of the dipolar Hamiltonian [18]:

$$\mathcal{H}_Z(t') = \sum_{i,j} b_{ij}(t') (2I_{iZ} I_{jZ} - I_{iX} I_{jX} - I_{iY} I_{jY}). \quad (5)$$

In the case of the hard RF pulse ($\omega_1 \gg \|\mathcal{H}_Z(t')\|$), the elements of the interaction Hamiltonian $\mathcal{H}_Z(t')$ noncommuting with I_Y may be ignored in Eq. (5) and the Liouville superoperator $\hat{L}_1(t')$ may be replaced by [21, 22]:

$$\hat{L}_1(t') = \left[-\omega_1 I_Y - \frac{1}{2} \mathcal{H}_Y(t'), \dots \right], \quad (6)$$

where [19]

$$\mathcal{H}_Y(t') = \sum_{i,j} b_{ij}(t') (2I_{iY}I_{jY} - I_{iX}I_{jX} - I_{iZ}I_{jZ}). \quad (7)$$

Assuming the first pulse is a 90° -pulse ($\omega_1 t_1 = \pi/2$), we have from Eqs. (1)–(7):

$$\rho(t_1) = \beta \left\{ 1 + \frac{i}{2} \int_0^{t_1} \hat{L}_Y(t') dt' - \frac{1}{8} \int_0^{t_1} dt'' \int_0^{t_1} \hat{L}_Y(t'') \hat{L}_Y(t') dt' + \dots \right\} I_X, \quad (8)$$

where

$$\hat{L}_Y(t') = [\mathcal{H}_Y(t'), \dots]. \quad (9)$$

After the first RF pulse the free evolution of the density operator is described by the Hamiltonian $\mathcal{H}_Z(t')$ and at the time τ (the time τ is measured from the beginning of the first pulse) the density operator has the following form:

$$\rho(\tau, t_1) = \hat{U}_0(\tau, t_1) \rho(t_1), \quad (10)$$

where the evolution superoperator $\hat{U}_0(\tau, t_1)$ is

$$\hat{U}_0(\tau, t_1) = 1 - i \int_{t_1}^{\tau} \hat{L}_Z(t') dt' - \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_{t_1}^{\tau} \hat{L}_Z(t'') \hat{L}_Z(t') dt' + \dots, \quad (11)$$

and

$$\hat{L}_Z(t') = [\mathcal{H}_Z(t'), \dots]. \quad (12)$$

Inserting Eq. (8) into Eq. (10) and with Eq. (11) we obtain:

$$\begin{aligned} \rho(\tau, t_1) = \beta \left\{ 1 + \frac{i}{2} \int_0^{t_1} \hat{L}_Y(t') dt' - \frac{1}{8} \int_0^{t_1} dt'' \int_0^{t_1} \hat{L}_Y(t'') \hat{L}_Y(t') dt' - i \int_{t_1}^{\tau} \hat{L}_Z(t') dt' \right. \\ \left. + \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_0^{t_1} \hat{L}_Z(t'') \hat{L}_Y(t') dt' - \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_{t_1}^{\tau} \hat{L}_Z(t'') \hat{L}_Z(t') dt' + \dots \right\} I_X. \quad (13) \end{aligned}$$

If at the time τ the second RF pulse is applied along the OX -axis in the rotating frame, the density operator becomes:

$$\rho(t_2, \tau, t_1) = \hat{U}_2(\tau + t_2, \tau) \rho(\tau, t_1), \quad (14)$$

where

$$\hat{U}_2(\tau + t_2, \tau) = 1 - i \int_{\tau}^{\tau+t_2} \hat{L}_2(t') dt' - \frac{1}{2} \int_{\tau}^{\tau+t_2} dt'' \int_{\tau}^{\tau+t_2} \hat{L}_2(t'') \hat{L}_2(t') dt' + \dots \quad (15)$$

and the Liouville superoperator $\hat{L}_2(t')$ has the form:

$$\hat{L}_2(t') = [-\omega_1 I_X + \mathcal{H}_Z(t'), \dots]. \quad (16)$$

In Eq. (14) and Eq. (15) t_2 is the width of the second RF pulse.

In the case of the hard RF pulse we may again retain only the secular terms or the terms commuting with I_X in $\mathcal{H}_Z(t')$, and replace $\hat{L}_2(t')$ by

$$\hat{L}_2(t') = \left[-\omega_1 I_X - \frac{1}{2} \mathcal{H}_X(t'), \dots \right], \quad (17)$$

where

$$\mathcal{H}_X(t') = \sum_{i,j} b_{ij}(t') (2I_{iX} I_{jX} - I_{iY} I_{jY} - I_{iZ} I_{jZ}). \quad (18)$$

Assuming the second pulse is also a 90°-pulse ($\omega_1 t_2 = \pi/2$) we obtain from Eqs. (13)–(17):

$$\begin{aligned} \rho(t_2, \tau, t_1) = & \beta \left\{ 1 + \frac{i}{2} \int_0^{t_1} \hat{L}_Z(t') dt' - \frac{1}{8} \int_0^{t_1} dt'' \int_0^{t_1} \hat{L}_Z(t'') \hat{L}_Z(t') dt' - i \int_{t_1}^{\tau} \hat{L}_Y(t') dt' \right. \\ & + \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_0^{t_1} \hat{L}_Y(t'') \hat{L}_Z(t') dt' - \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_{t_1}^{\tau} \hat{L}_Y(t'') \hat{L}_Y(t') dt' \\ & - \frac{1}{4} \int_{\tau}^{\tau+t_2} dt'' \int_0^{t_1} \hat{L}_X(t'') \hat{L}_Z(t') dt' \\ & \left. + \frac{1}{2} \int_{\tau}^{\tau+t_2} dt'' \int_{t_1}^{\tau} \hat{L}_X(t'') \hat{L}_Y(t') dt' + \dots \right\} I_X. \quad (19) \end{aligned}$$

After the second RF pulse the free evolution of the density operator is described by the Liouville superoperator $\hat{L}_Z(t')$ (Eq. (12)) and at time t (the time t is measured from the beginning of the first pulse) the density operator has the form:

$$\rho(t, t_2, \tau, t_1) = \hat{U}_0(t, \tau + t_2) \rho(t_2, \tau, t_1), \quad (20)$$

where

$$\hat{U}_0(t, \tau + t_2) = 1 - i \int_{\tau+t_2}^t \hat{L}_Z(t') dt' - \frac{1}{2} \int_{\tau+t_2}^t dt'' \int_{\tau+t_2}^t \hat{L}_Z(t'') \hat{L}_Z(t') dt' + \dots \quad (21)$$

Inserting Eq. (19) into Eq. (20) and with Eq. (21) we obtain:

$$\begin{aligned}
\rho(t, t_2, \tau, t_1) = \beta & \left\{ 1 + \frac{i}{2} \int_0^{t_1} \hat{L}_Z(t') dt' - \frac{1}{8} \int_0^{t_1} dt'' \int_0^{t_1} \hat{L}_Z(t'') \hat{L}_Z(t') dt' - i \int_{t_1}^{\tau} \hat{L}_Y(t') dt' \right. \\
& + \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_0^{t_1} \hat{L}_Y(t'') \hat{L}_Z(t') dt' - \frac{1}{2} \int_{t_1}^{\tau} dt'' \int_{t_1}^{\tau} \hat{L}_Y(t'') \hat{L}_Y(t') dt' \\
& - \frac{1}{4} \int_{\tau}^{\tau+t_2} dt'' \int_0^{t_1} \hat{L}_X(t'') \hat{L}_Z(t') dt' + \frac{1}{2} \int_{\tau}^{\tau+t_2} dt'' \int_{t_1}^{\tau} \hat{L}_X(t'') \hat{L}_Y(t') dt' \\
& + \frac{1}{2} \int_{\tau+t_2}^t dt'' \int_0^{t_1} \hat{L}_Z(t'') \hat{L}_Z(t') dt' - \int_{\tau+t_2}^t dt'' \int_{t_1}^{\tau} \hat{L}_Z(t'') \hat{L}_Y(t') dt' \\
& \left. - \frac{1}{2} \int_{\tau+t_2}^t dt'' \int_{\tau+t_2}^t \hat{L}_Z(t'') \hat{L}_Z(t') dt' \dots \right\} I_X. \quad (22)
\end{aligned}$$

The observed transient response of the ensemble of spins or the two-pulse signal is given by [18–20]:

$$V(t, t_2, \tau, t_1) = \frac{\text{Tr} \{ \overline{\rho(t, t_2, \tau, t_1)} I_X \}}{\text{Tr} (I_X^2)}, \quad (23)$$

where the upper bar denotes the average of the density operator on the random motions of nuclei.

Substituting Eq. (22) into Eq. (23) and evaluating the traces in Eq. (23) we obtain:

$$V(t, t_2, \tau, t_1) = \beta \left\{ 1 - \frac{1}{2} F(t, t_2, \tau, t_1) + \dots \right\}. \quad (24)$$

Here

$$\begin{aligned}
F(t, t_2, \tau, t_1) = & \frac{1}{4} \int_0^{t_1} dt'' \int_0^{t_1} h(t'', t') dt' + \int_{t_1}^{\tau} dt'' \int_0^{t_1} h(t'', t') dt' \\
& + \int_{t_1}^{\tau} dt'' \int_{t_1}^{\tau} h(t'', t') dt' - \int_{\tau+t_2}^t dt'' \int_0^{t_1} h(t'', t') dt' \\
& - 2 \int_{\tau+t_2}^t dt'' \int_{t_1}^{\tau} h(t'', t') dt' + \int_{\tau+t_2}^t dt'' \int_{\tau+t_2}^t h(t'', t') dt', \quad (25)
\end{aligned}$$

and

$$h(t'', t') = W \sum_{i,j} \overline{b_{ij}(t'') b_{ij}(t')} \quad (26)$$

is the correlation function of the dipolar local fields [18–20].

In Eq. (26) [18–20]

$$W = \frac{3}{4} \gamma^4 \hbar^2 I(I+1) \frac{1}{N}, \quad (27)$$

$$b_{ij}(t') = R_{ij}^{-3}(t') [1 - 3 \cos^2 \Theta_{ij}(t')]. \quad (28)$$

In order to calculate the correlation function $h(t'', t')$ we consider the simple model of the molecular motion in solids between discrete lattice sites Ω_k ($k = 1, 2, \dots, n$) [23]. We assume that the random process describing the molecular motions in solids is a stationary Markovian process [24, 25]. For a stationary Markovian process the correlation function $h(t'', t')$ depends only on $|t'' - t'|$ and $h(t)$ ($t > 0$) can be written as [24, 25]:

$$h(t) = W \sum_{i,j} \sum_{l,m} P(\Omega_l) P(\Omega_l, 0 | \Omega_m, t) b_{ij}(\Omega_l) b_{ij}(\Omega_m). \quad (29)$$

Here $P(\Omega_l)$ is the probability that at time $t = 0$, the random function $b_{ij}(t)$ is equal to $b_{ij}(\Omega_l)$, while $P(\Omega_l, 0 | \Omega_m, t)$ is the conditional probability that if at time $t = 0$ the random function will be equal to $b_{ij}(\Omega_m)$ [24, 25].

For the random Markovian process the conditional probability $P(\Omega_l, 0 | \Omega_m, t)$ satisfies the Smoluchowski equation [24, 25]

$$\frac{\partial}{\partial t} P(\Omega_l, 0 | \Omega_m, t) = \sum_{k=1}^n P(\Omega_l, 0 | \Omega_k, t) W_{km}, \quad (30)$$

with the conditions [24, 25]

$$P(\Omega_l, 0 | \Omega_m, 0) = \delta_{lm}, \quad (31)$$

$$\sum_{m=1}^n P(\Omega_l, 0 | \Omega_m, t) = 1, \quad (32)$$

$$\sum_{m=1}^n W_{lm} = 0. \quad (33)$$

In Eq. (30) W_{km} ($k \neq m$) is the rate constant which describes the probability of the random variable $\Omega(t)$ changing from Ω_k to Ω_m by one jump [24, 25].

Assuming that ($l \neq m$)

$$W_{lm} = \frac{1}{n \tau_c} \quad (34)$$

it is easily obtained from Eq. (30):

$$P(\Omega_l, 0 | \Omega_m, t) = \frac{1}{n} \left[1 - \exp\left(-\frac{t}{\tau_c}\right) \right] + \exp\left(-\frac{t}{\tau_c}\right) \delta_{lm}. \quad (35)$$

Inserting Eq. (35) into Eq. (29) and assuming that $P(\Omega_l) = 1/n$ we have

$$h(|t|) = \overline{M}_2 + \Delta M_2 \exp\left(-\frac{|t|}{\tau_c}\right), \quad (36)$$

where

$$\overline{M}_2 = W \sum_{i,j} \left[\frac{1}{n} \sum_{k=1}^n b_{ij}(\Omega_k) \right]^2 \equiv W \sum_{i,j} (\overline{b}_{ij})^2 \quad (37)$$

is the second moment of motionally narrowed NMR line [18, 23] and

$$\Delta M_2 = M_2 - \overline{M}_2. \quad (38)$$

In Eq. (38)

$$M_2 = W \sum_{i,j} b_{ij}^2 \quad (39)$$

is the second moment of NMR line in rigid lattice [18, 23].

Using the correlation function (36) and calculating the integrals in Eq. (25) we obtain the following expression for the solid-echo signal:

$$V(t, t_2, \tau, t_1) = \beta \left\{ 1 - \frac{1}{2} \overline{M}_2 \left[t - \left(2\tau + t_2 - \frac{t_1}{2} \right) \right]^2 - \Delta M_2 \tau_c^2 R(t, t_2, \tau, t_1, \tau_c) + \dots \right\}, \quad (40)$$

where

$$\begin{aligned} R(t, t_2, \tau, t_1, \tau_c) = & -\frac{7}{4} + \frac{t}{\tau_c} - \frac{3t_1}{4\tau_c} - \frac{t_2}{\tau_c} - \frac{1}{4} \exp\left(-\frac{t_1}{\tau_c}\right) - \exp\left(-\frac{t_2}{\tau_c}\right) \\ & - \frac{1}{2} \exp\left(-\frac{t}{\tau_c}\right) + \frac{1}{2} \exp\left(-\frac{\tau - t_1}{\tau_c}\right) - \frac{1}{2} \exp\left(-\frac{t - t_1}{\tau_c}\right) \\ & + \frac{1}{2} \exp\left(-\frac{\tau + t_2}{\tau_c}\right) + \exp\left(-\frac{t - \tau}{\tau_c}\right) + \exp\left(-\frac{t - \tau - t_2}{\tau_c}\right) \\ & + \frac{1}{2} \exp\left(-\frac{\tau}{\tau_c}\right) + \frac{1}{2} \exp\left(-\frac{\tau + t_2 - t_1}{\tau_c}\right). \end{aligned} \quad (41)$$

In the case of the polycrystalline sample we must average Eq. (40) over all possible orientations of the crystallites. If we denote the averaged values of $\overline{M_2}$ and ΔM_2 as $\langle \overline{M_2} \rangle$ and $\langle \Delta M_2 \rangle$, then for the small τ and t we may write:

$$\begin{aligned}
 V(t, t_2, \tau, t_1) &= \beta \left\{ 1 - \frac{1}{2} \langle \overline{M_2} \rangle \left[t - \left(2\tau + t_2 - \frac{t_1}{2} \right) \right]^2 - \langle \Delta M_2 \rangle \tau_c^2 R(t, t_2, \tau, t_1, \tau_c) + \dots \right\} \\
 &\approx \frac{\hbar \omega}{kT} \exp \left\{ -\frac{1}{2} \langle \overline{M_2} \rangle \left[t - \left(2\tau + t_2 - \frac{t_1}{2} \right) \right]^2 \right. \\
 &\quad \left. - \langle \Delta M_2 \rangle \tau_c^2 R(t, t_2, \tau, t_1, \tau_c) \right\}. \tag{42}
 \end{aligned}$$

From Eq. (41) and Eq. (42) it follows that at $\langle M_2 \rangle \tau_c^2 \gg 1$ (the case of rigid lattice) and at $\langle M_2 \rangle \tau_c^2 \ll 1$ (the case of motionally narrowed NMR line) the maximum of solid-echo signal is observed at $t_e = 2\tau + t_2 - t_1/2$ [15–17].

If we consider the delta-functions approximation of the RF pulses and put in Eq. (41) $t_1 = t_2 = 0$ we obtain from Eq. (42) the known result [7]. From the analysis of Eq. (42) it follows that in the slow-motion regime ($\langle M_2 \rangle \tau_c^2 \approx 1$) the amplitude of the echo signal is reduced and the maximum of the echo signal is shifted to the end of the second pulse.

3 Results and Discussion

We apply the obtained theoretical results to the analysis of temperature dependences of the solid-echo signals in polycrystalline benzene (C_6H_6) and polycrystalline ammonium chloride (NH_4Cl). These compounds were chosen because they have been the subject of extensive studies by different experimental NMR techniques (see references in [26, 27]). It is now well-established that in NH_4Cl there are the reorientations of the ammonium ions about threefold and twofold symmetry axes [26]; and in solid benzene there are the reorientations of C_6H_6 molecules about its sixfold symmetry axis [27].

All 1H experiments were performed on an NMR spectrometer operating at 60 MHz. The length of $\pi/2$ pulse was 3.6 μs . The temperature ranges studied were from 133 to 273 K for NH_4Cl and from 100 to 278 K for C_6H_6 . Measurements below 100 K were not possible with the equipment applied.

Experimental measurements of the maximum echo time position (t_e) at different pulse spacing for polycrystalline NH_4Cl and C_6H_6 as function of temperature are shown in Figs. 1 and 2. One can see that for any pulse spacing the maximum of two-pulse signal does not depend on the temperature in the region $T > 160$ K for NH_4Cl and in the region $T > 130$ K for C_6H_6 and maximum echo is observed at $t_e \approx 2\tau + t_2 - t_1/2$.

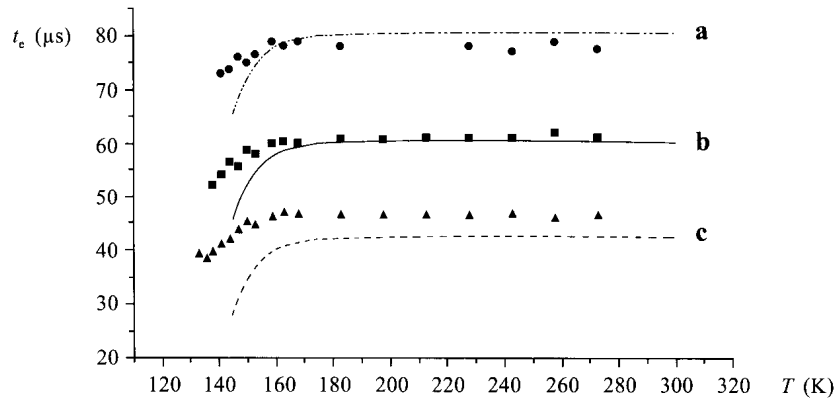


Fig. 1. Temperature dependence of the time position of the maximum solid-echo amplitude for polycrystalline NH_4Cl at different pulse spacings: **a** $\tau = 39.4 \mu\text{s}$; **b** $\tau = 29.4 \mu\text{s}$; **c** $\tau = 20.4 \mu\text{s}$.

At $T < 160 \text{ K}$ for NH_4Cl and $T < 130 \text{ K}$ for C_6H_6 the time positions of the maximum of the echo signals are shifted to the end of the second pulse and echo signals disappear. The solid and broken lines in Figs. 1 and 2 are the theoretical curves for the time positions of the echo maximum, as calculated from Eq. (42) with the results of relaxation studies of these compounds [14, 26–28]. For NH_4Cl we used the following parameters: $\tau_c = (2.16 \cdot 10^{-14} \text{ s}) \cdot \exp(19.85 \text{ kJ} \cdot \text{mol}^{-1}/RT)$; $\overline{M}_2 = 4.74 \cdot 10^{-8} \text{ T}^2$; $\Delta M_2 = 46.15 \cdot 10^{-8} \text{ T}^2$. For C_6H_6 we used $\tau_c = (1.3 \cdot 10^{-14} \text{ s}) \cdot \exp(17.5 \text{ kJ} \cdot \text{mol}^{-1}/RT)$, $\overline{M}_2 = 1.5 \cdot 10^{-8} \text{ T}^2$; $\Delta M_2 = 7.16 \cdot 10^{-8} \text{ T}^2$. The agreement between theory and experiment is reasonable especially since no parameters have been adjusted.

Experimental measurements of the maximum echo amplitude at two different pulse spacings for polycrystalline NH_4Cl and C_6H_6 as function of temperature

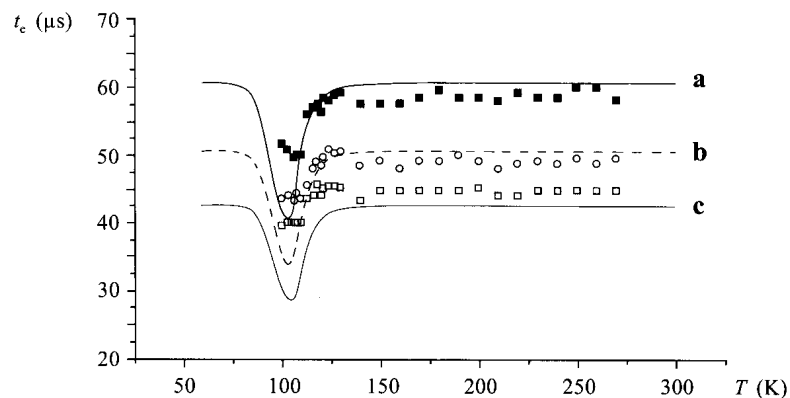


Fig. 2. Temperature dependence of the time position of the maximum solid-echo amplitude for polycrystalline C_6H_6 at different pulse spacings: **a** $\tau = 29.4 \mu\text{s}$; **b** $\tau = 24.4 \mu\text{s}$; **c** $\tau = 20.4 \mu\text{s}$.

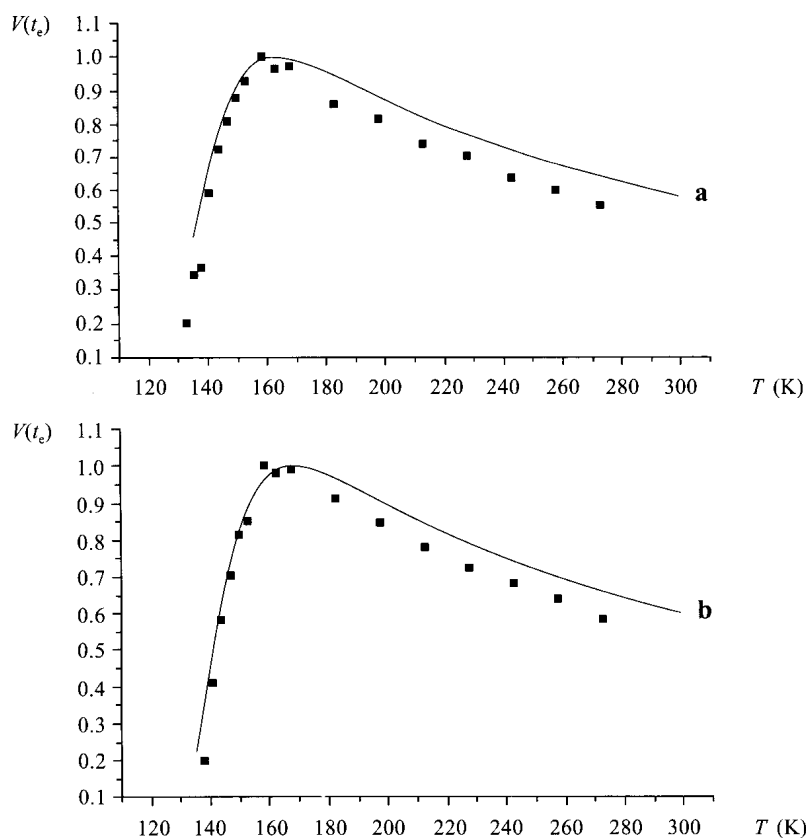


Fig. 3. Temperature dependence of the maximum solid-echo amplitude for polycrystalline NH_4Cl at different pulse spacings: **a** $\tau = 20.4 \mu\text{s}$; **b** $\tau = 29.4 \mu\text{s}$.

are shown in Figs. 3 and 4. In all cases, the plotted points were normalized so that the maximum value of the echo amplitude was set equal to 1.0. From Figs. 3 and 4 it follows that for different pulse intervals the maximum echo amplitude increases and then falls down rapidly to reach minimum values in the region of 130 K for NH_4Cl and 100 K for C_6H_6 . The solid lines in Figs. 3 and 4 are the theoretical curves obtained from Eq. (42) with the same parameters as for the theoretical curves in Figs. 1 and 2. From Figs. 3 and 4 we see that the developed theory describes rather well the observed temperature dependences of the echo amplitude.

In conclusion, it is clear that the temperature dependences of the time position and the amplitude of the solid-echo are sensitive in certain temperature regions to the details and thermal parameters of molecular dynamic process.

The observed difference between theory and experiment may be connected with the fact that the results of our calculations are sensitive to the used parameters. Another explanation of this discrepancy is that the developed theory is only

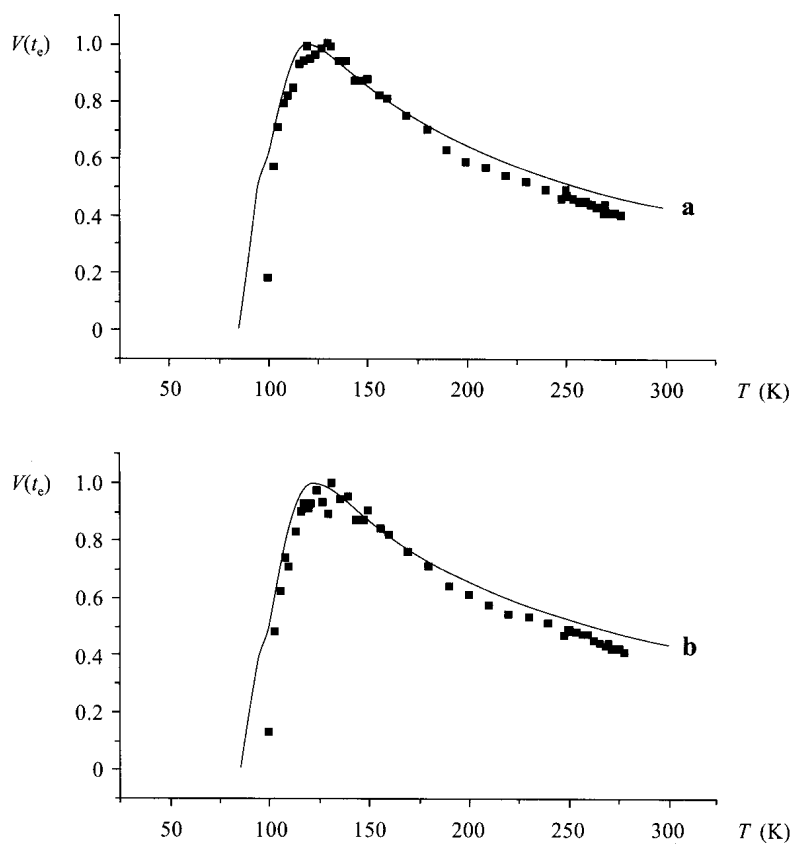


Fig. 4. Temperature dependence of the maximum solid-echo amplitude for polycrystalline C_6H_6 at different pulse spacings: **a** $\tau = 20.4 \mu s$; **b** $\tau = 24.4 \mu s$.

applied when $\tau_c^{-1} \ll \omega_1, \omega_0$ (secular approximation). In order to understand these discrepancies we intend to extend our study of solid echoes in solids with molecular mobility using the other compounds and considering the nonsecular contributions to the relaxation of the solid-echo signals.

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