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SPIN ECHO AMPLITUDE IN PULSED NMR: EFFECTS OF ELECTRIC QUADRUPOLE INTERACTION

by

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ABSTRACT

In ferromagnetic systems where the NMR spectra present unresolved electric quadrupole interactions the spin echo amplitude shows an oscillatory behaviour as a function of the time separation τ between excitation pulses. These oscillations are in general attenuated.

In the present work spin echo amplitudes are numerically computed, using a simple algorithm. The calculation is made for several values of the nuclear spin I and for different excitation conditions. The curves are compared with available analytical calculations. The amplitudes of the multiple spin echoes (for $I > 1$) are also obtained.

The calculation is extended to the case of non-axially symmetric electric field gradients (EFG's) and to inhomogeneous EFG's; the attenuation in the corresponding amplitudes confirms that these conditions explain curves observed experimentally.

Key-words: Pulsed NMR; Spin echo; Quadrupole interaction; Ferromagnetic systems.

INTRODUCTION

A spin echo is formed when an ensemble of magnetic moments in an inhomogeneous magnetic field is excited by a sequence of short radio-frequency pulses (E.L. Hahn (1)). The echo is the signal arising from the re-focussing of the precessing magnetic moments; in Hahn's experiment two pulses are applied, separated by a time interval τ, and the echo appears at $t = 2\tau$ (Fig. 1). The formation of a spin echo in a nuclear magnetic resonance (NMR) experiment was discussed by Hahn, using Bloch's equations (1).

Solomon (2) examined the case of nuclei with spin $I = 5/2$ in the presence of a magnetic field and an electric field gradient (EFG). Assuming an interaction with the rf field much larger than the electric quadrupole interaction, several echoes result. For a sequence of two pulses, one turning the spins by an angle $\theta = \pi/2$, the second by an angle $\theta' = \pi/5$, Solomon obtained multiple echoes, i.e., four more echoes at $3\tau/2$, $5\tau/2$, 3τ and 4τ, besides Hahn's echo at $t = 2\tau$. A density matrix formalism was used; the Hamiltonian included a magnetic term and an electrostatic term (interaction with a distribution of axially symmetric EFG's). The effect of the rf pulses was introduced via rotation operators.

Butterworth (3) considered both a distribution of magnetic fields and of EFG's in the Hamiltonian and computed, for $I = 3/2$ and $I = 5/2$, the dependence of the echo amplitude on the turning angle of the second pulse. Rotation operators accounted for the effect of the rf pulses.

In ferromagnetic systems where the NMR spectrum presents unresolved electric quadrupole interactions, the echo amplitudes versus pulse separation τ show an oscillatory behaviour. The oscillations can be interpreted as beats arising from the unequal spacing of the nuclear hyperfine levels. They have been computed analytically for $I = 3/2$ by Abe et al. (4) within a perturbation scheme, for the case where the hyperfine interaction is either smaller or larger than the interaction with the rf pulses.

We adopt in the present work the model described in ref. (4), but in order to calculate the spin echo amplitudes we develop an algorithm which allows numerical computation for any value of the nuclear spin and for different excitation conditions (i.e. duration of the pulses, intensity of the rf field). In principle the computation is free from any perturbative approximation. In next Section the main steps in the treatment of ref. (4) are summarized and an algorithm to compute numerically the spin echo amplitudes for $t = 2\tau$, 3τ, etc. is presented. In sequence several applications of this algorithm are carried out. A comparison of the numerical method with the analytical results of ref. (4) is given. Other applications are detailed, particularly the derivation of multiple echoes and the oscillations in the curve of echo amplitude versus τ. Finally, effects of asymmetry and inhomogeneity in the EFG are discussed.

FORMULATION OF THE PROBLEM

A spin system consisting of one nuclear species of magnetic moment μ and electric quadrupole moment Q interacts with an internal magnetic field and an electric field gradient. The spin system is submitted to two radio-frequency pulses of duration t_w and t'_w, separated by a time interval τ, as sketched in the diagram shown in Fig. 1; five regions 0, I, II, III and IV are then defined.

In a frame of reference that rotates with an angular frequency w, identical to the rf frequency and near the Larmor frequency w_o, the Hamiltonian of the hyperfine interaction (magnetic and electric) is given by

$$\mathcal{H}_{hf} = (w_o - w) I^z + a (I^z)^2 \quad , \tag{1}$$

where $a = 3e^2 qQ/4I(2I-1)$ is the electric quadrupole interaction parameter.

The time dependent part of the Hamiltonian is given by the interaction of the spin system with the rf:

$$\mathcal{H}_{Int} = \begin{cases} 0 & \text{in region } 0 \\ w_1 I^y & \text{" " I} \\ 0 & \text{" " II} \\ w'_1 I^y & \text{" " III} \\ 0 & \text{" " IV} \end{cases}$$

where w_1 is the angular frequency of the nuclear precession induced by the radio-frequency.

One assumes an inhomogeneous distribution of hyperfine fields given by a function $f(w_o)$, centred in w_{oo} and of width δw_o.

As a consequence the average value of the transverse component of the nuclear magnetization, in the rotating frame, is given by

$$S'(t) = \int dw_o F(w_o) s'(t) \tag{3}$$

where s'(t) is obtained from

$$s'(t) = \mathrm{Tr}\left[\sigma'(t) I^+\right] \tag{4}$$

and $\sigma'(t)$, the density matrix operator, is given by the equation of motion

$$i d\sigma'(t)/dt = \left[\mathcal{H}' , \sigma'(t)\right] \tag{5}$$

I^+ is the transverse component of the spin operator and $\mathcal{H}'$ is the complete Hamiltonian in the rotating frame.

A formal solution of Eq. (5) is

$$\sigma'(t) = U_{IV} U_{III} U_{II} U_I \sigma'(0) U_I^\dagger U_{II}^\dagger U_{III}^\dagger U_{IV}^\dagger \tag{6}$$

where

$$U_i = \exp\left[A_i\right] \ , \ i = I, \ldots, IV$$

$$A_I = - i t_w \left[(w_o - w) I^z + a(I^z)^2 + w_1 I^y)\right]$$

$$A_{II} = - i(\tau - t_w) \left[(w_o - w) I^z + a(I^z)^2\right]$$

U_{III} and U_{IV} are similar to U_I and U_{II}, where t_w is replaced by t'_w , w_1 by w'_1 and $\tau - t_w$ by $t - \tau - t'_w$; $\sigma'(0)$ is the solution of the density matrix in thermodynamic equilibrium $(t = 0)$.

In order to compute $s'(t)$ for $t = n\tau$, where n is an integer and τ is the interval between rf pulses, we remind that $F(w_o)$ is the convolution of the distribution of Larmor frequencies $f(w'_o)$ and the Lorentzian lineshape $h(w'_o)$ of natural linewidth, that is

$$F(w_o) = \int dw'_o f(w'_o) h(w'_o) \tag{7}$$

Finally we want to obtain $S(t)$ at $t = n\tau$, where $S(t)$ is the average of the transverse component of the nuclear spin in the laboratory system

$$S(t) = \exp(iwt) S'(t) \tag{8}$$

The combined application of Eqs. (4), (6), (7) and (8) allows us to obtain explicitly the spin echo amplitudes at $t = 2\tau$ (Hahn's echo), but also at 3τ, etc. As a consequence of the distribution of hyperfine fields the average of the echo amplitude in the laboratory system is given by

$$E_1(2\tau - t_w + t'_w) = \exp\left[-(2\tau - t_w + t'_w)/T_2\right] \mathrm{Tr}\left[\tilde{\sigma}(2\tau - t_w + t'_w) I^+\right] \tag{9}$$

where T_2 is the transverse relaxation time, and

$$\tilde{\sigma}(2\tau - t_w + t'_w) = U_{IV} U_{III} U_{II} \tilde{\sigma}(t_w) U^{\dagger}_{II} U^{\dagger}_{III} U^{\dagger}_{IV} \tag{10}$$

and $\tilde{\sigma}(t_w)$ has non-zero elements only on the line one above the diagonal. These elements are those of $\sigma'(t_w)$. This result may be generalized for multiple echoes: one has to compute the echo amplitudes $t=3\tau$, etc.; now $\tilde{\sigma}(t_w)$ has non-zero elements on line two above the diagonal, three above, etc.

The above outlined prescription to obtain the echo amplitudes was applied by Abe et al. (4) to the case of $I=3/2$. Analytical solutions were obtained, choosing appropriate eigenstates of I^z (or I^y). Expression 6 was then explicitly computed using the identity

$$\exp(\beta B)\exp\left[-\beta(B+C)\right] = 1 - \int_0^\beta d\lambda \exp(\lambda B) C \exp\left[-\lambda(B+C)\right] \tag{11}$$

$$\exp\left[-\beta(B+C)\right] = \exp(-\beta B)\{1 - \int_0^\beta d\lambda \exp(\lambda B) C \exp(-\lambda B)$$
$$+ \int_0^\beta d\lambda_1 \int_0^{\lambda_1} d\lambda_2 \exp(\lambda_1 B) C \exp\left[-(\lambda_1-\lambda_2)B\right] C \exp(-\lambda_2 B) + \ldots\} \tag{12}$$

where B and C are operators and B does not commute with C, but C contains a perturbative parameter which allows a cutoff in the above expansion.

In the present work we perform numerical computations of the echo amplitudes using a procedure which is not perturbative. To evaluate $\sigma'(t_w)$, $\tilde{\sigma}(2\tau)$ and finally $E_1(2\tau)$, expressions 9 and 10, we make use of the following identity (5).

$$U_i = M_i U(D_i) M_i^{-1} \tag{13}$$

where D_i is a diagonal matrix built with the eigenvalues of A_i; M_i, the modal matrix, is constructed with the eigenvalues associated to the eigenvalues of A_i. The following quantities are investigated: a) the amplitude of the first spin echo as a function of the time interval τ between the rf pulses, for the cases $I = 1$ and $I = 3/2$ using both the analytical methodology of Abe et al. (4) and the algorithm developed in this work; b) echo amplitudes versus angle of rotation (θ') of the second rf pulse for different values of nuclear spin; c) echo amplitudes versus separation τ between pulses, for different excitation conditions; d) amplitudes of the echoes of order 2, 3, etc; e) effects of asymmetry in the electric field gradient and of a distribution of hyperfine interactions on the amplitude of the echoes.

The numerical methodology described here is illustrated for the case $I = 1$ and a sequence of $\pi/2$ and π pulses, in Appendix A.

NUMERICAL COMPUTATION OF SPIN ECHOES

We have chosen a set of parameters based on experimental results of ^{59}Co NMR in intermetallic compounds of formula RCo_2 (R is a rare-earth) (Hirosawa and Nakamura (6)). The numerical results obtained in the computation of echo amplitudes with these parameters are presented below.

a) Comparison with the analytical results

With the parameters $T = 4.2$ K, $w_0 = 60$ MHz, $a = 0.6$ MHz, $t_w = 0.5\ \mu s$, $t_w' = 1.0\ \mu s$, we have computed numerically, for $I = 1$, the amplitude of the first echo ($n = 1$) as a function of τ. This was done for two different angles of rotation of the second pulse ($\theta' = \pi$ and $\theta' = \pi/2$). These curves are shown in Fig. 2a and 2b, together with the plot of the expression given by Abe et al. (4). In an analogous way, the case $I = 3/2$, $\theta' = \pi$ was studied (Fig. 2c).

A visual inspection of figures 2a, 2b and 2c reveals the following features: i) $I = 1$: the amplitudes of the oscillations derived numerically and analytically show a very good agreement. It should be noted that for $\theta' = \pi/2$ the oscillations are above the axis of τ, but for $\theta' = \pi$ the echo amplitudes oscillate above and below zero; ii) $I = 3/2$: there is a close agreement between the curves obtained by the two methods: however, a small phase difference is apparent. The amplitudes show a small excursion below the axis (for $\theta' = \pi$), and the oscillation frequency is different from the case $I = 1$.

b) Echo amplitudes versus angle θ'

We have also applied the numerical method to compute the echo amplitudes for different values of the angle of rotation of the second pulse (θ'). The echo amplitude for a fixed τ and for different values of w_1' and t_w' (cases $I = 1$, 3/2, 2, 5/2, 3 and 7/2, are given in the series of curves in Fig. 3a and 3b.

c) Echo amplitudes versus separation τ

The echo heights versus separation τ between the two pulses, for the angle θ' that gives maximum echoes are shown in Fig. 4, for different values of spin I. The behaviour of the echoes versus τ for different values of the turning angle θ' and for nuclear spin $I = 5/2$ is given in Fig. 5; here the duration of the two pulses was 0.5 μs and 1.0 μs, respectively, and the rf intensity of the second pulse (w_1') was varied. The presence of higher harmonics is evident; this effect had also been observed experimentally (4,7).

d) Echoes of order $n > 1$

The numerical method allows us to verify the occurrence of multiple echoes and to compute the amplitudes $E_1(2\tau)$, $E_2(3\tau)$ and so on. These amplitudes have been calculated for $I = 3/2$, 5/2 and 7/2 and are shown in Fig. 6 for $I = 3/2$.

e) Effect of asymmetry and inhomogeneity of the EFG

The spin echo oscillations observed experimentally in some ferromagnetic systems show attenuation in the amplitude, as a function of the pulse separation τ. Degani and Kaplan (8) suggested that this attenuation could arise from a non-axially symmetric EFG (asymmetry parameter $\eta \neq 0$). The expression of the electrostatic part of the nuclear hyperfine Hamiltonian is, in this case:

$$\mathcal{H}_Q = a\left[(I^z)^2 - I(I+1)/3 + ((I^+)^2 + (I^-)^2)\eta/6\right] \qquad (14)$$

We have, in our calculation, compared the amplitude for a non-axial EFG to the axial case, for I = 1, 3/2, 5/2 and 7/2, for two values of the angle of rotation of the second pulse θ'. Some results are shown in figures 7a and 7b. The computed echo amplitudes can be described by the following function:

$$E_1(2\tau) = \exp(-2\tau/T_2)\sum_{n=0}^{2I-1} C_n \cos(n\eta\tau)\cos\left[2na\tau + \delta_n\right] \qquad (15)$$

The curves $E_1(2\tau)$ computed for $\eta \neq 0$ allow us the conclusion that the asymmetry may be responsible for the echo attenuation. For I = 1 (Fig. 7a) an apparent attenuation is observed in the calculated curves for $\eta = 0.2$.

In the situation where the resonant nuclei experience different electric field gradients, a distribution of values of the parameter a has to be introduced in Eq. (3):

$$S'(t) = \int dw_o F(w_o) s'(t) f(a) da \qquad (16)$$

where f(a) is taken as a normalized gaussian. The amplitude of the oscillations of the first echo for I = 1 ($\theta' = \pi/2$ and π) and I = 3/2 ($\theta' = \pi/2$) computed with a distribution of EFG's is shown in Fig. 8a, 8b and 8c for a distribution of the para meter a 0.1 MHz wide. The amplitudes are attenuated and this result is obtained for axially symmetric and non-axially symmetric field gradients. The damping of quadrupolar oscillations has been related to the width of the distribution of quadrupolar frequencies $\Delta\nu_Q$ in the analysis of ^{11}B ($I = 3/2$) NMR by Erdmann et al. (9). They have fitted their experimental data to the function

$$E_1(2\tau) = f(2\tau)\{ C_0 + \exp(-4\pi\Delta\nu_Q\tau)\left[C_1\cos(2\pi\bar{\nu}_Q\tau + \delta_1) + C_2\cos(4\pi\bar{\nu}_Q\tau + \delta_2)\right]\} \qquad (17)$$

where $f(2\tau)$ contains the T_2 decay and $\bar{\nu}_Q = \bar{a}/\pi$.

CONCLUSIONS

We have shown that a wide range of nuclear spin echo phenomena can be described within a simple model, originally proposed by Abe et al. (4); the numerical method here developed allows the computation of echoes as a function of various relevant parameters and is free from any perturbative approximation that may restrict the useful range of model parameters.

The effect of the interaction with electric field gradients can be taken into account, and the resulting oscillations computed for different excitation conditions. It emerges from such analysis that the damping of these oscillations results naturally either from the inhomogeneity of the EFG's or from the asymmetry of the EFG.

APPENDIX A

In the case of $I = 1$, the matrices A_i ($i = 0$, I, II, III, IV) are constructed using eigenstates of I^z

$$A_0 = -\frac{\hbar(\omega_0 + a)}{k_B T}\begin{bmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \frac{a - \omega_0}{\omega_0 + a} \end{bmatrix} \tag{A1}$$

$$A_I = \begin{bmatrix} -it_w(a + \Delta\omega) & -w_1 t_w/\sqrt{2} & 0 \\ w_1 t_w/\sqrt{2} & 0 & -w_1 t_w/\sqrt{2} \\ 0 & w_1 t_w/\sqrt{2} & -it_w(a - \Delta\omega) \end{bmatrix} \tag{A2}$$

$$A_{II} = i(\tau - t_w)\begin{bmatrix} a + \Delta\omega & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & a - \Delta\omega \end{bmatrix} \tag{A3}$$

$$A_{III} = \begin{bmatrix} -it'_w(a + \Delta\omega) & -w'_1 t'_w/\sqrt{2} & 0 \\ w'_1 t'_w/\sqrt{2} & 0 & -w'_1 t'_w/\sqrt{2} \\ 0 & w'_1 t'_w/\sqrt{2} & -it'_w(a - \Delta\omega) \end{bmatrix} \tag{A4}$$

$$A_{IV} = -i(t-\tau-t'_w)\begin{bmatrix} a+\Delta\omega & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & a-\Delta\omega \end{bmatrix} \qquad (A5)$$

For a sequence of $\pi/2$ and π pulses ($w_i t_w = \pi/2$ and $w'_i t'_w = \pi$) and a = 0.6 MHz, $T = 4.2$ K, $t'_w = 2t_w = 1\mu s$, $w = w_0$, the matrices A_I, M_I and $U(D_I)$ (for example), are approximately given by

$$A_I = \begin{bmatrix} -0.3i & -1.1 & 0 \\ 1.1 & 0 & -1.1 \\ 0 & 1.1 & 0.3i \end{bmatrix} \qquad (A6)$$

$$M_I = \begin{bmatrix} 1.0 & 1.0 & 1.0 \\ -1.6i & 0 & 1.3i \\ -1.0 & 1.0 & -1.0 \end{bmatrix} \qquad (A7)$$

$$U(D_I) = \begin{bmatrix} 0.14+0.99i & 0 & 0 \\ 0 & 0.95-0.29i & 0 \\ 0 & 0 & 0.16-0.99i \end{bmatrix} \qquad (A8)$$

Also,

$$\sigma(0) = \frac{e^{A_o}}{Tr\left[e^{A_o}\right]} = \frac{\underline{1}}{3} + 10^{-5}\begin{bmatrix} -0.37 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0.36 \end{bmatrix} \qquad (A9)$$

where $\underline{1}$ is the unit matrix and

$$\tilde{\sigma}(t_w) = \begin{bmatrix} 0 & -0.25+0,04i & 0 \\ 0 & 0 & -0.25-0.04i \\ 0 & 0 & 0 \end{bmatrix} \qquad (A10)$$

In a similar way, from A_{II}, A_{III} and A_{IV} one obtains U_{II} U_{III} and U_{IV}.

From (10) one gets

$$\tilde{\sigma}(t=2\tau-t_w+t'_w)I^+ = \begin{bmatrix} 0 & -0.0036+0.0005i & 0.0175-0.0512i \\ 0 & 0.342-0.105i & 0 \\ 0 & 0 & 0.342+0.105i \end{bmatrix}$$

(A11)

and finally,

$$s'(t=2\tau-t_w+t'_w)\Big|_{\tau=0} \mathrm{Tr}\left[\tilde{\sigma}(2\tau-t_w+t'_w)\ I^+\right]\Big|_{\tau=0} = 0.684$$

and from (9)

$$E_1(2\tau-t_w+t_{w'})\Big|_{\tau=0} = 0.698$$

The numerical program repeats this task for several values of τ.

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FIGURE CAPTIONS

Fig. 1 - Time diagram showing the sequence of two rf pulses of width t_ω and t'_ω separated by a time interval τ, and the spin echo at $t = 2\tau$ (Hahn's echo). Five time regions are defined: 0, I, II, III and IV.

Fig. 2a - Amplitude of the first spin echo, as a function of the time interval τ between the rf pulses, for nuclear spin I = 1; the dashed curve is the numerical calculation and the points the analytical results (parameters a = 0.6 MHz, ω_o = 60 MHz, $\theta' = \pi$).

Fig. 2b - Same as Fig. 2a, for $\theta' = \pi/2$.

Fig. 2c - Echo amplitudes for I = 3/2 and $\theta' = \pi$.

Fig. 3a - Echo amplitudes versus angle of rotation of the second rf pulse for spin 1 (....), spin 3/2 (-.-.-) and spin 2 (---).

Fig. 3b - Echo amplitudes for I = 5/2 (....), I = 3 (-.-.-), I = 7/2 (---). The parameters are τ = 5.5 μs and t'_ω = 1 μs.

Fig. 4 - Echo amplitude versus separation τ, between pulses for I = 1 (....), I = 3/2 (-.-.-) and I = 2(---) for θ' giving maximum echoes. Parameters ω_o = 60 MHz, a = 0.6 MHz.

Fig. 5 - Echo amplitudes versus τ for I = 5/2 and different turning angles of the second pulse: $\theta' = \pi/3$ (....), $\theta' = \pi$ (-.-.-) and $\theta' = 2\pi$ (---). Parameters: ω_o = 60 MHz and a = 0.6 MHz.

Fig. 6 - First echo amplitude $E_1(2\tau)$ (....), second echo amplitude $E_2(3\tau)$ (-.-.-) and third echo $E_3(4\tau)$ (---), for $I = 3/2$.

Fig. 7a - Echo amplitude versus τ for $I = 1$, $\theta' = \pi$ and different values of the electric field gradient asymmetry parameter η: $\eta = 0.0$ (....), $\eta = 0.2$ (-.-.-) and $\eta = 0.4$ (---).

Fig. 7b - Echo amplitude for $I = 1$, $\theta' = \pi/2$ and different values of the EFG η: $\eta = 0.0$ (....), $\eta = 0,2$ (-.-.-) and $\eta = 0.4$ (---).

Fig. 8a - Echo amplitude versus τ for different values of the asymmetry parameter η, and a distribution of the quadrupole interaction parameter a of width 0.1 MHz, centred in $a_o = 0.6$ MHz. The parameters are $I = 1$, $\theta' = \pi$, $\eta = 0.0$ (....), $\eta = 0.2$ (-.-.-) and $\eta = 0.4$ (---). The attenuation of the echo amplitudes is clearly visible.

Fig. 8b - Echo amplitudes, for angle of rotation $\theta' = \pi/2$. The echo attenuation is also apparent.

Fig. 8c - Same for $I = 3/2$ and $\theta' = \pi/2$.

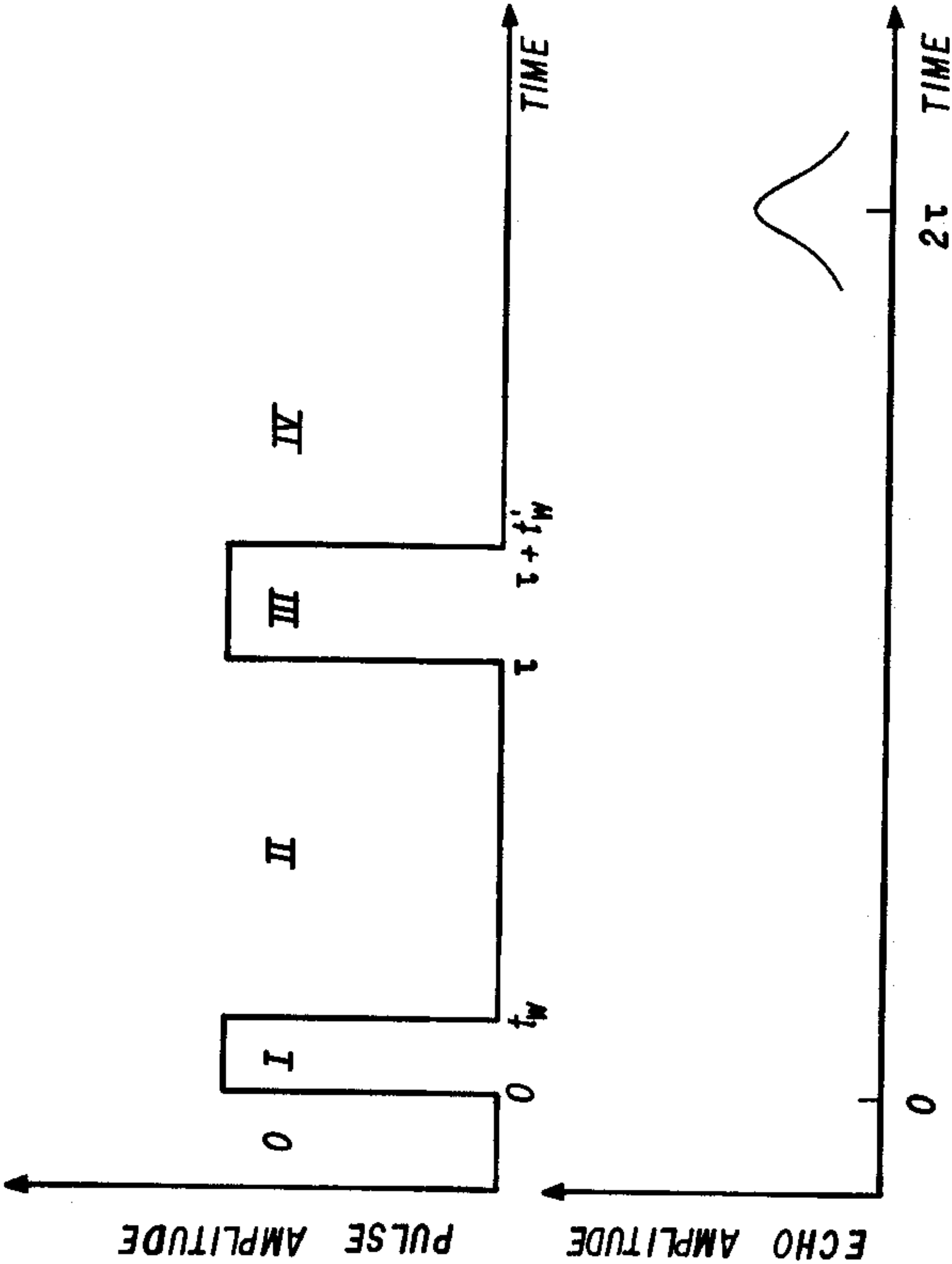
PULSE AMPLITUDE
ECHO AMPLITUDE
TIME
TIME
0
I
II
III
IV
0
t_w
τ
$\tau + t'_w$
0
2τ

FIG. 1

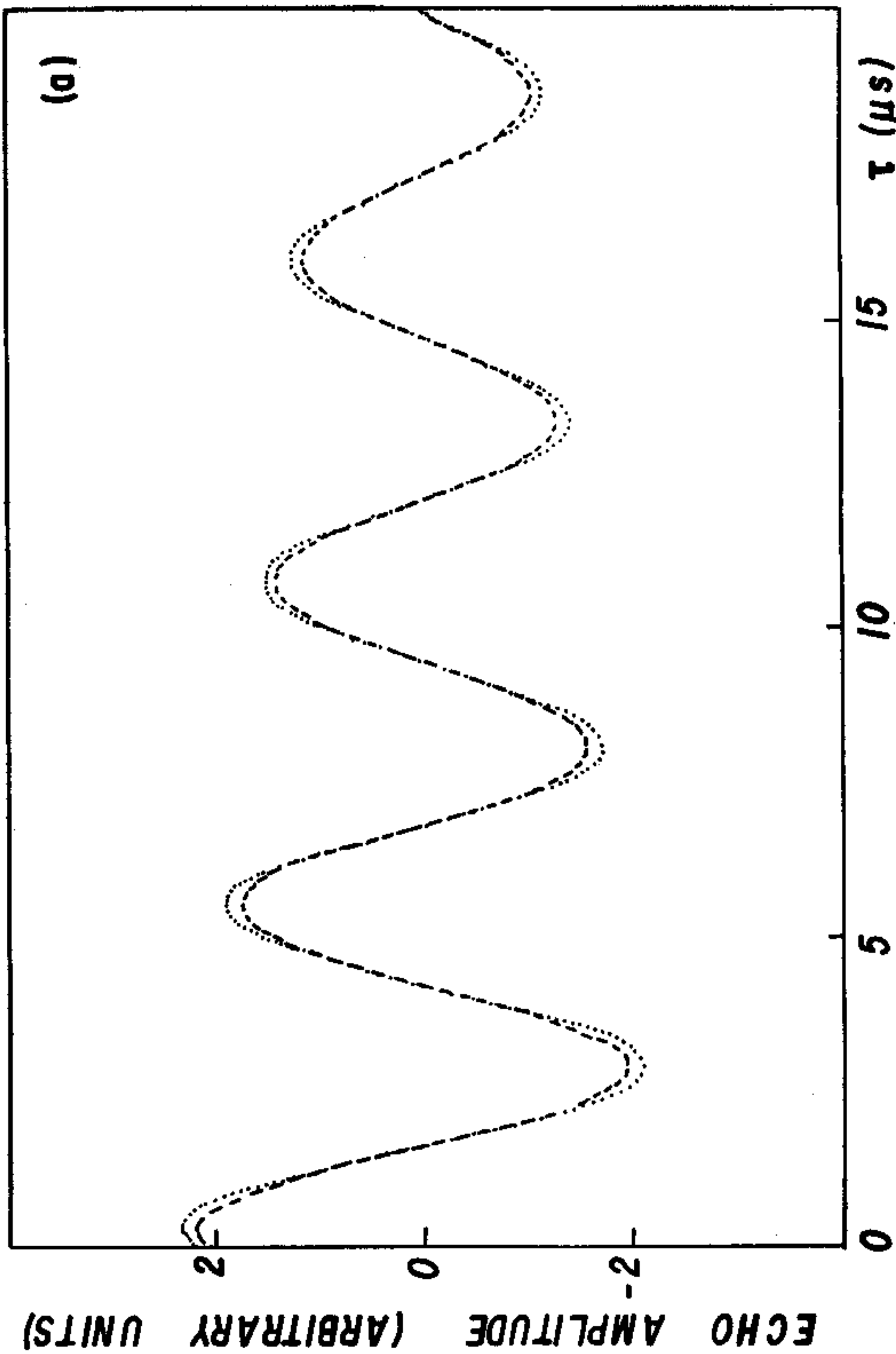

FIG. 2a

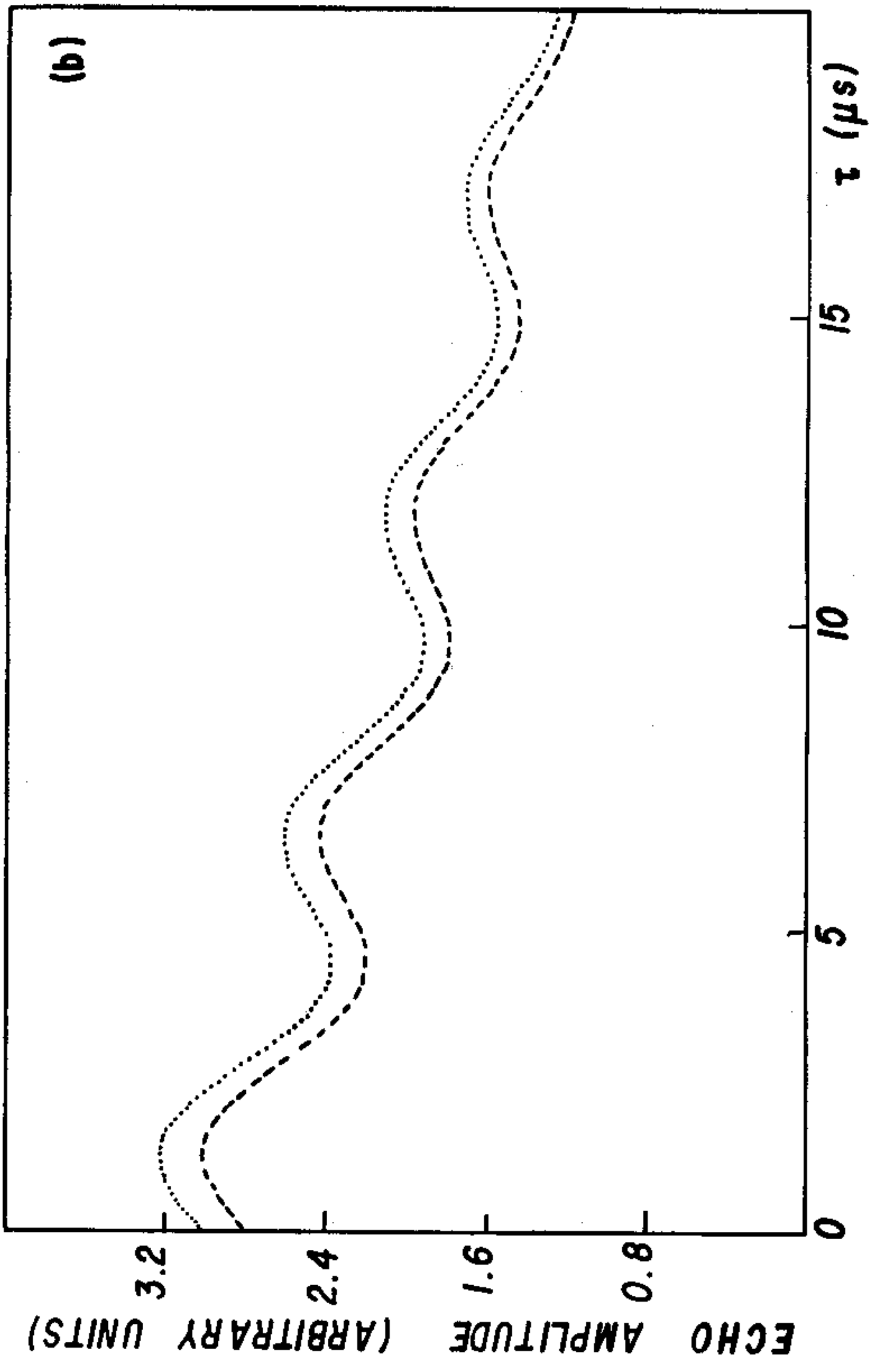

Fig. 2b

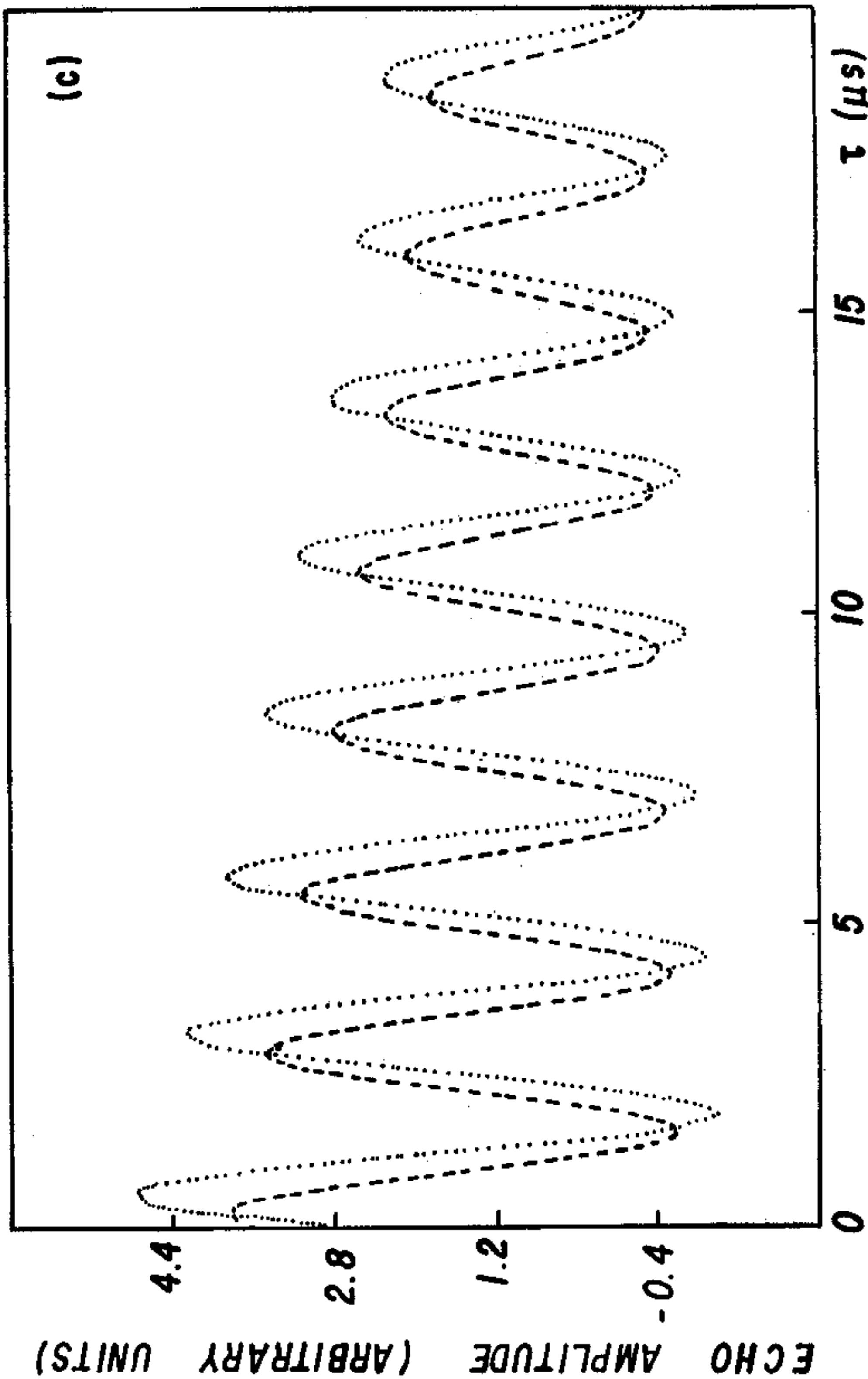

Fig. 2c

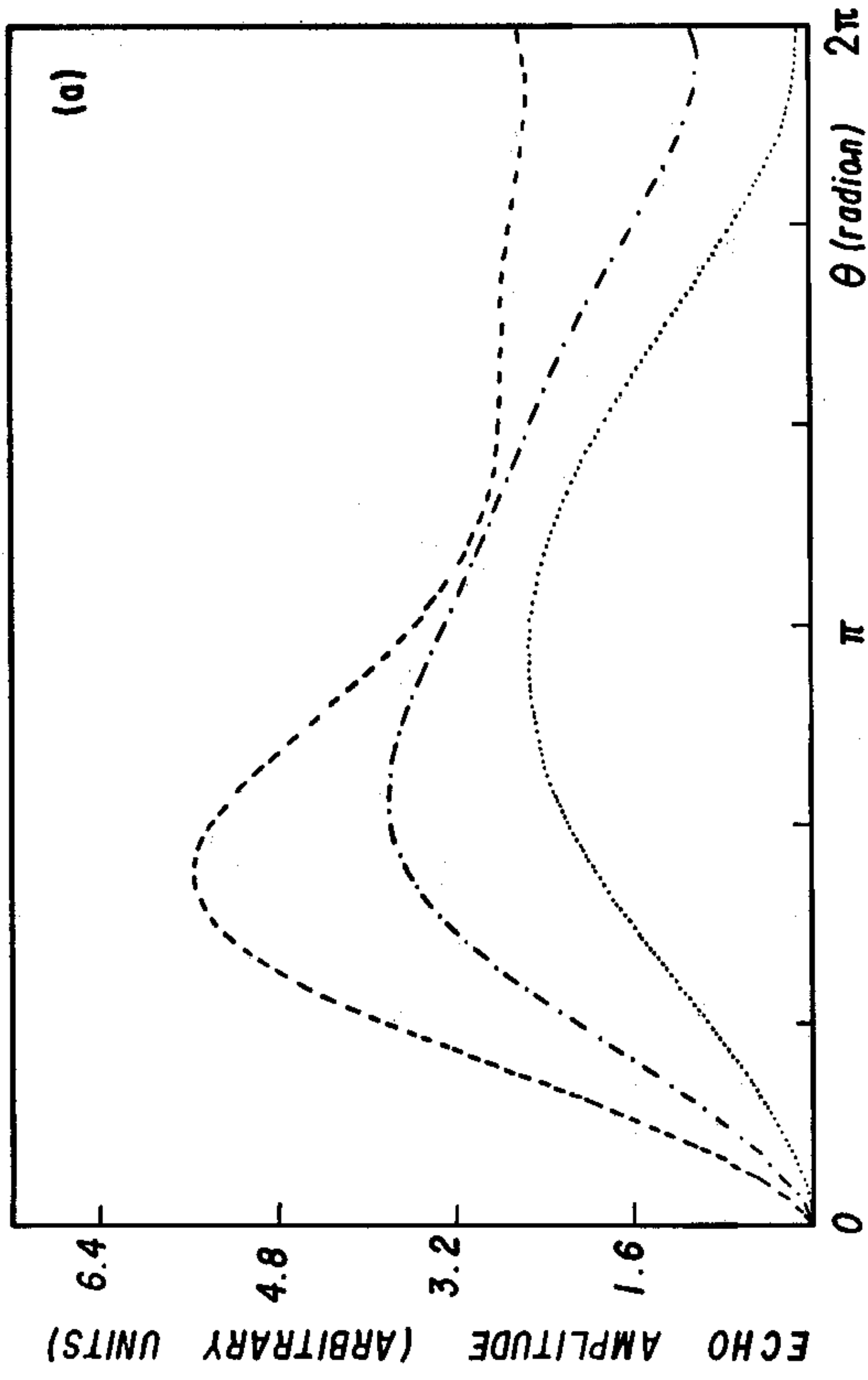

Fig. 3a

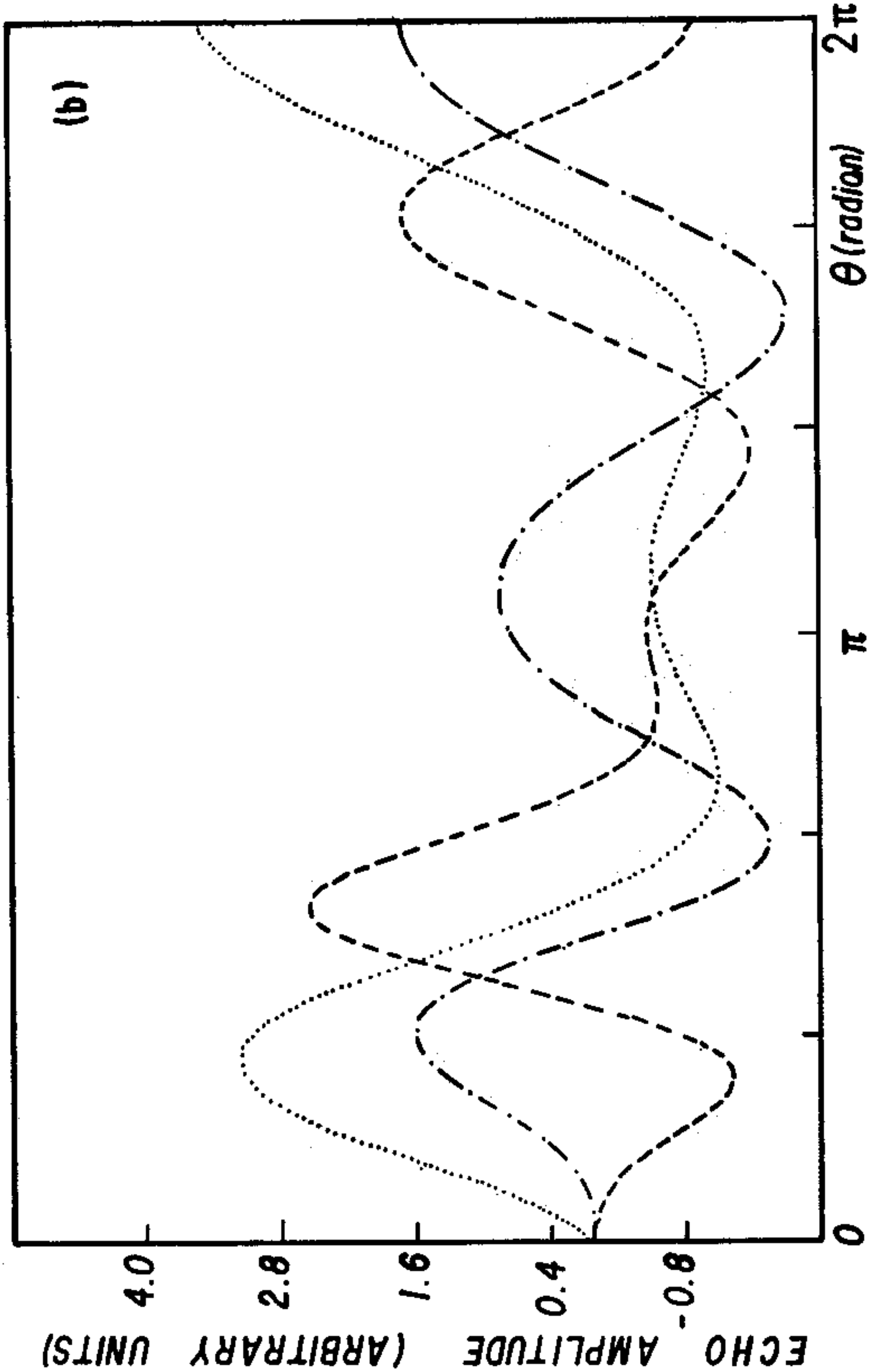

Fig. 3b

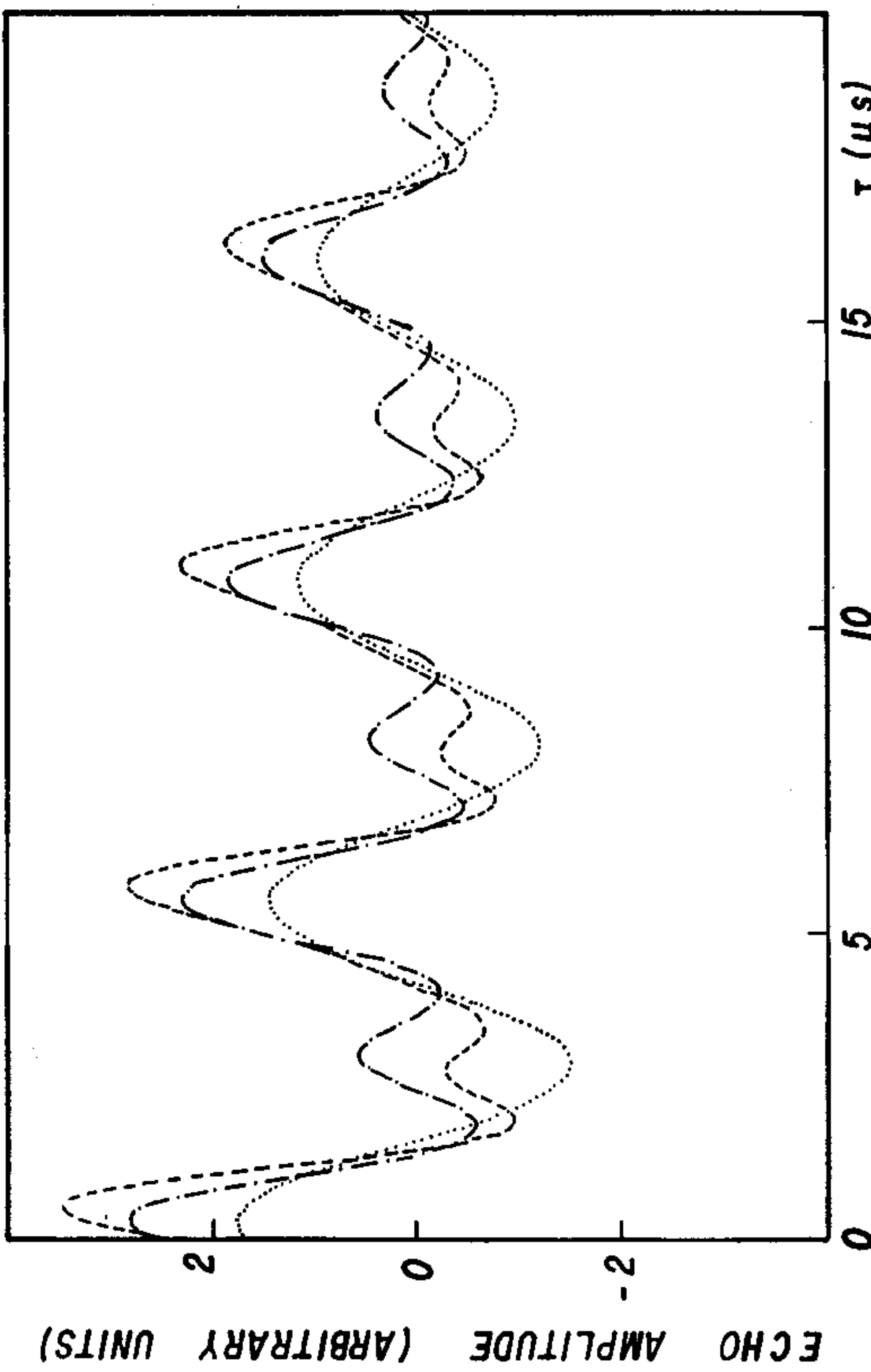

Fig. 4

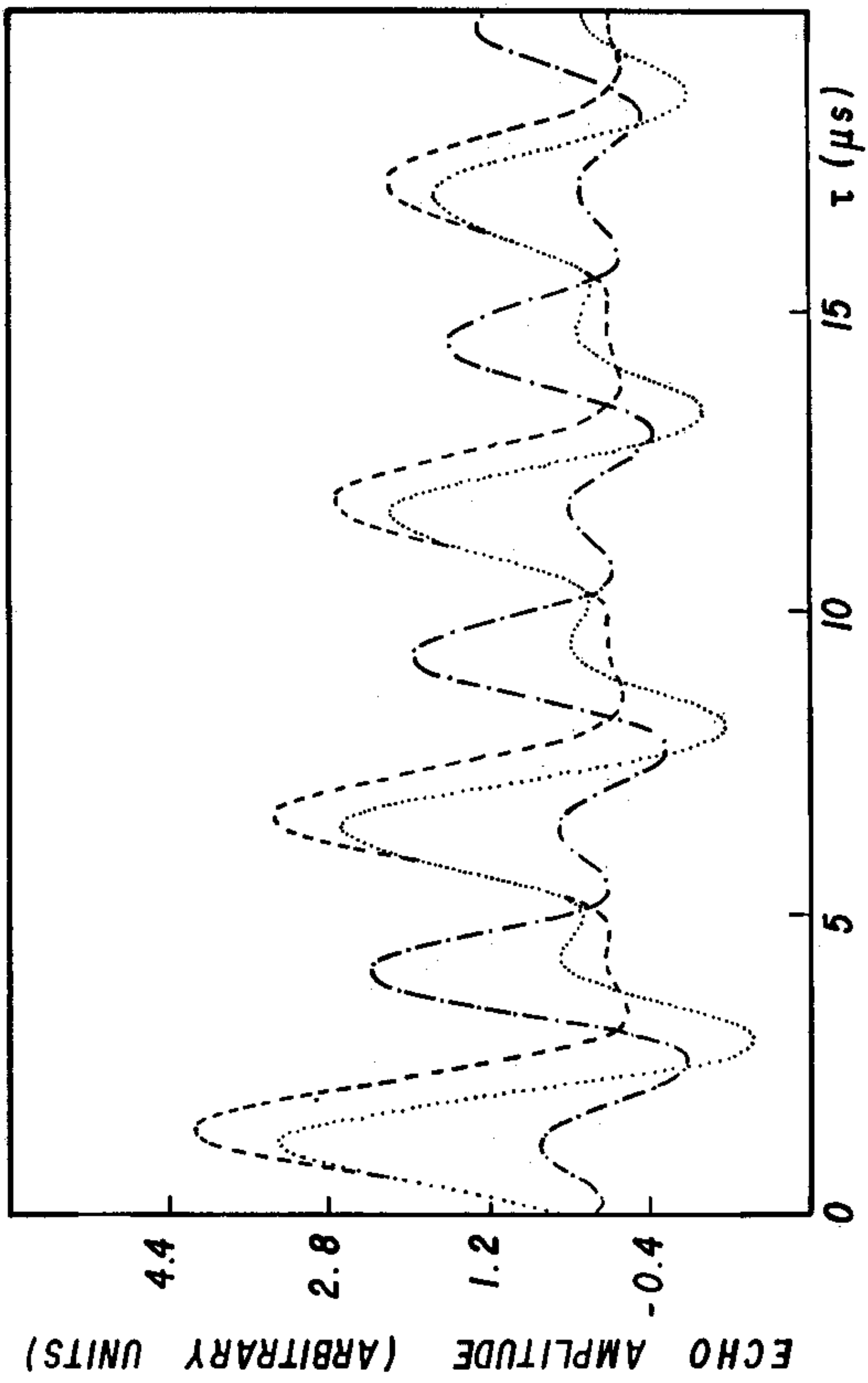

Fig. 5

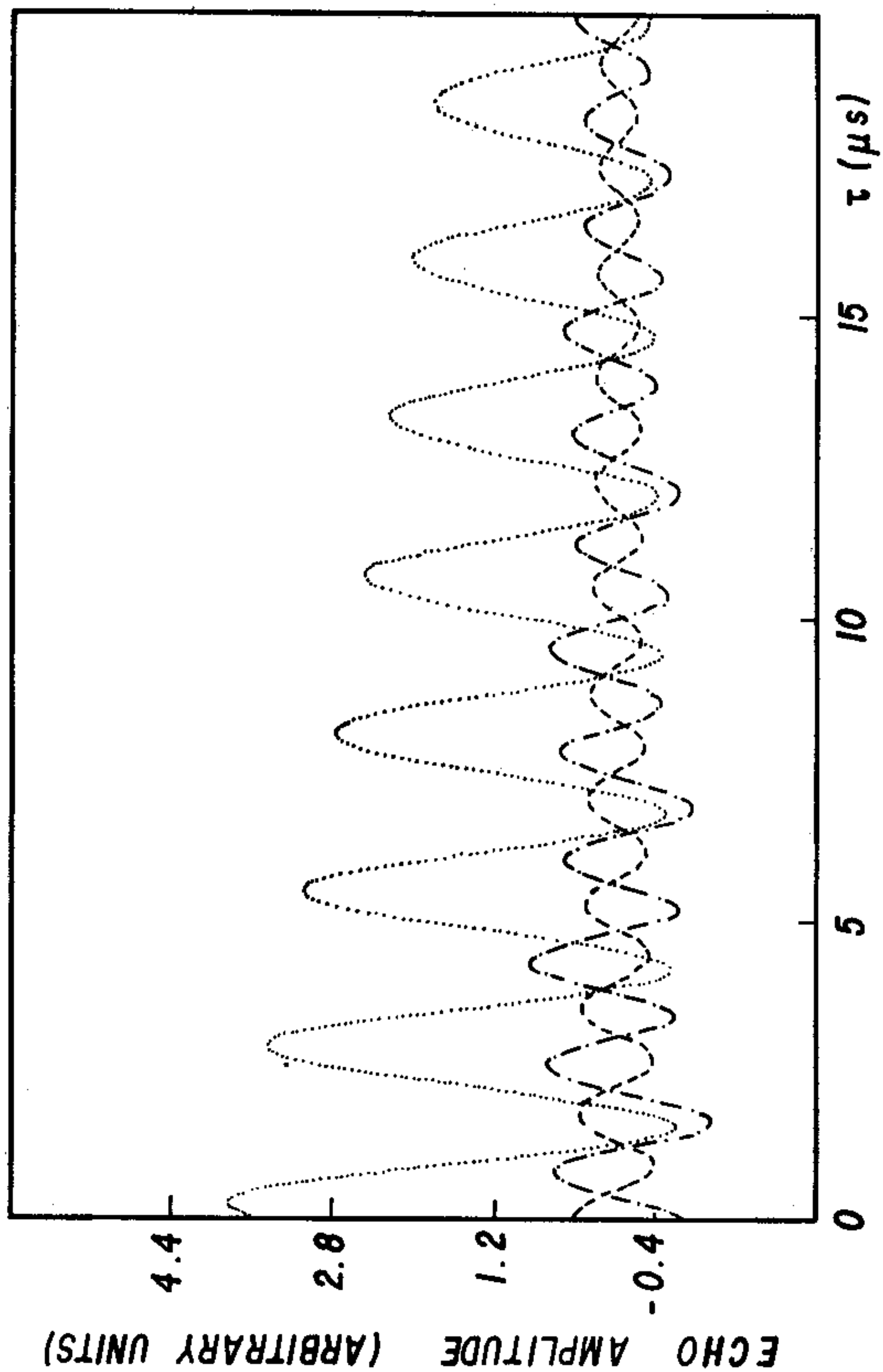

Fig. 6

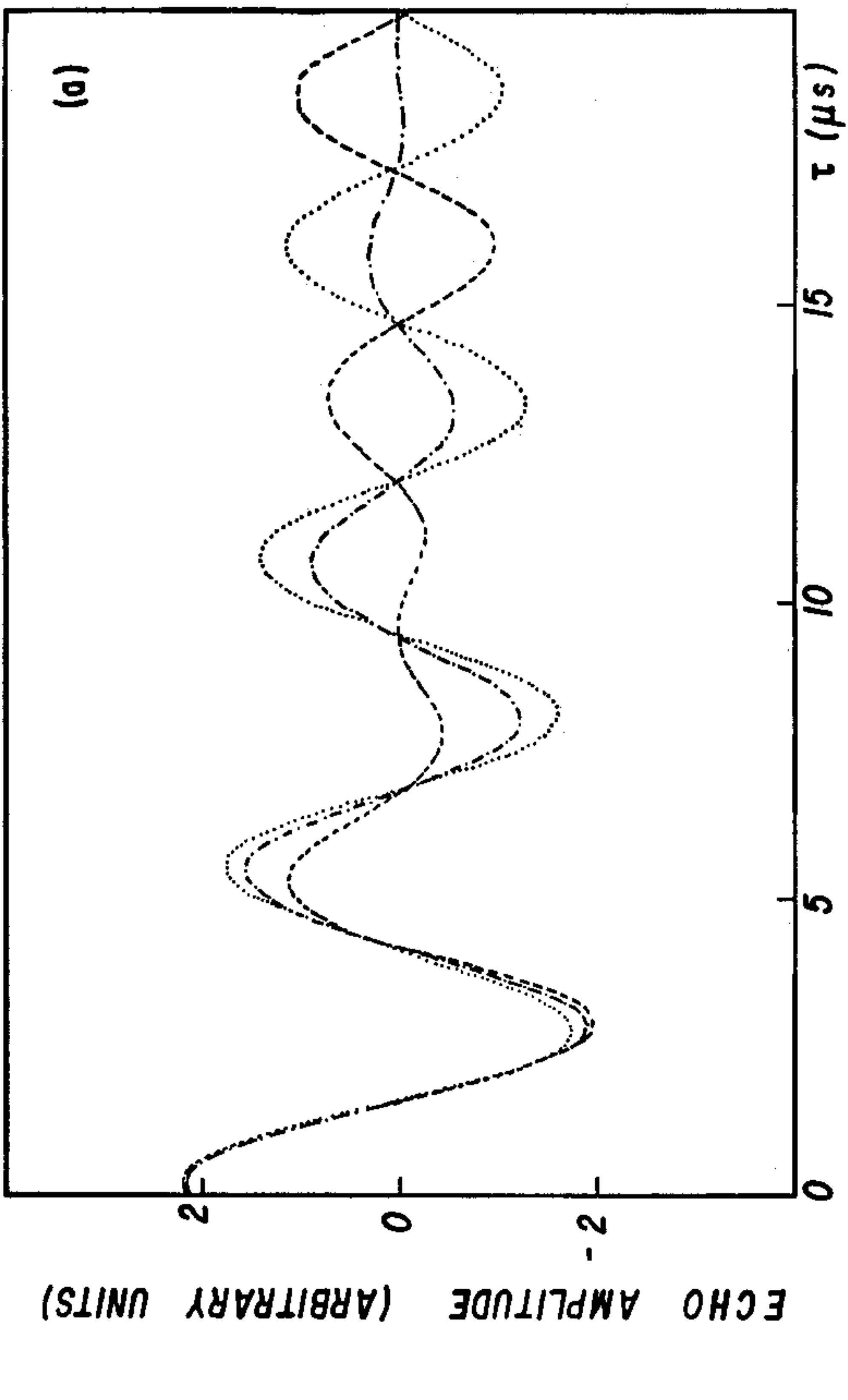

Fig. 7a

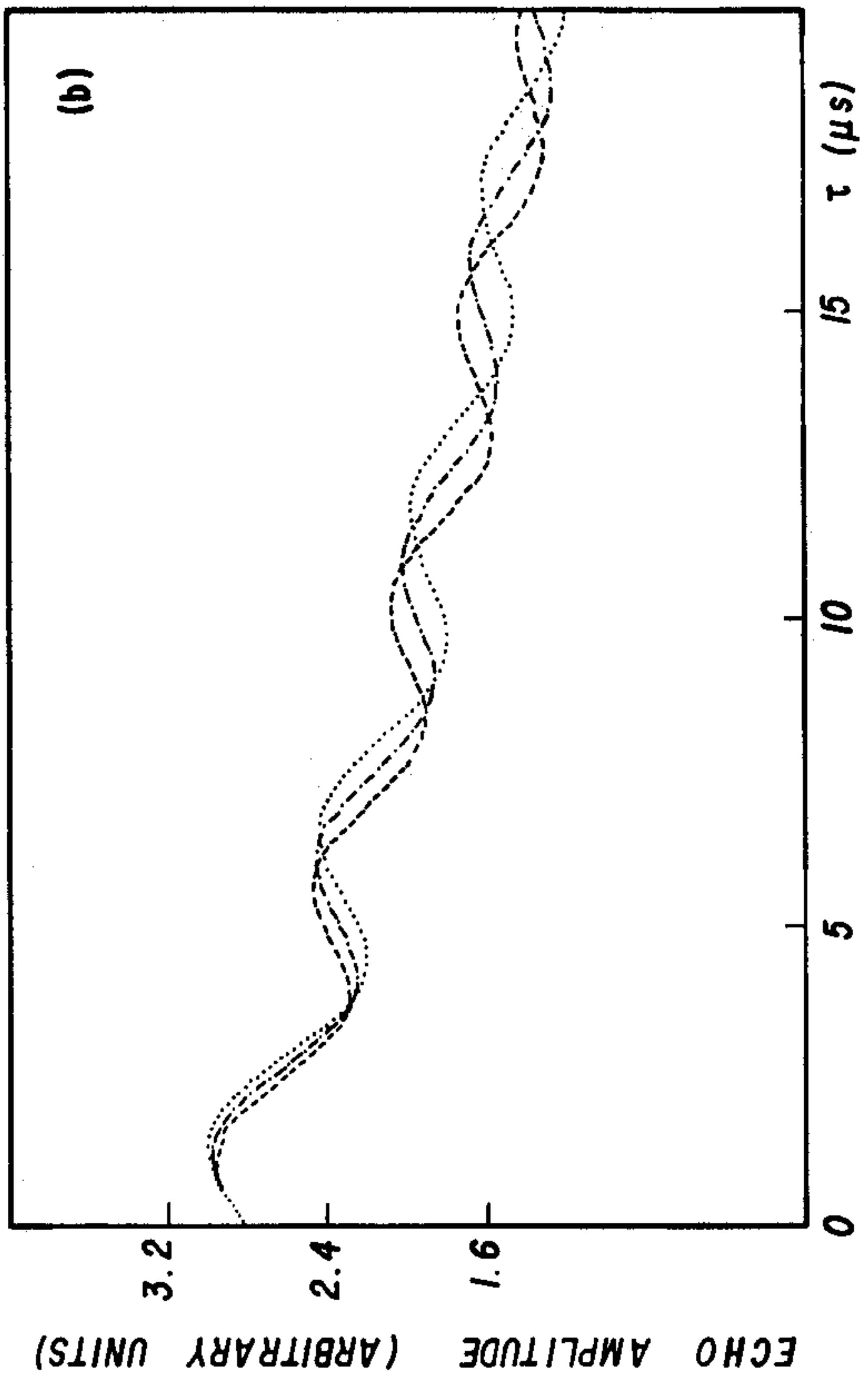

Fig. 7b

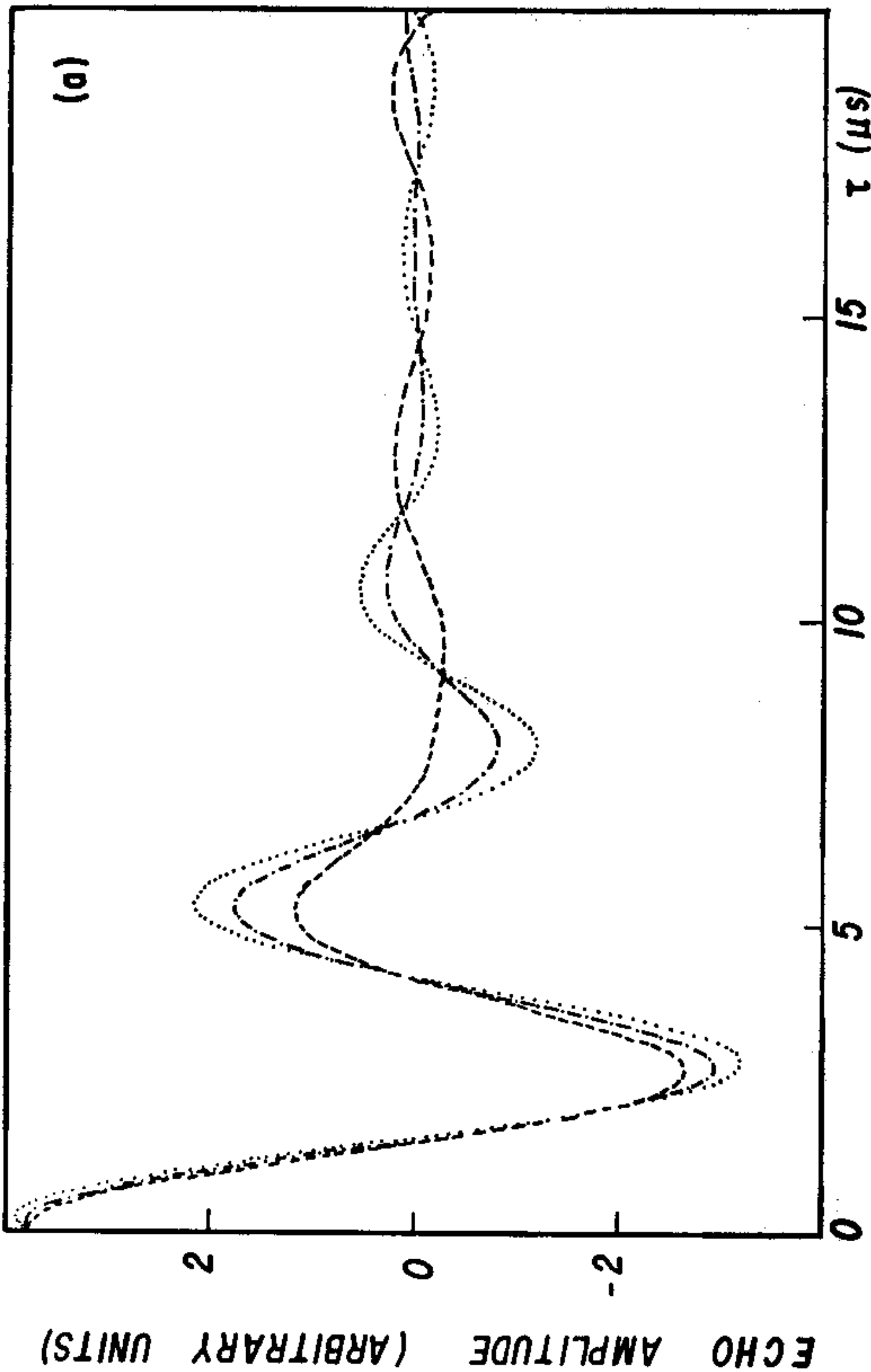

Fig. 8a

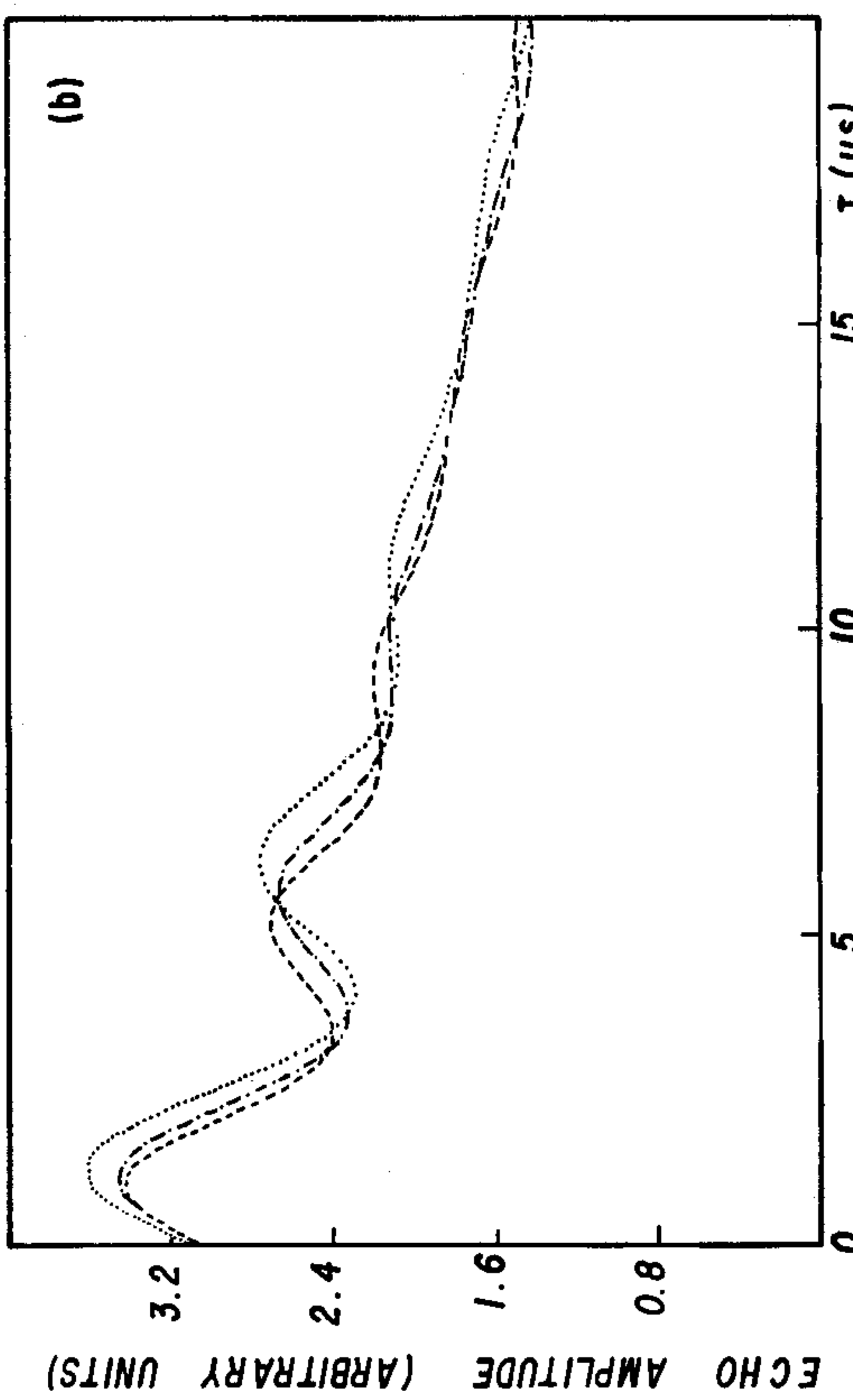

Fig. 8b

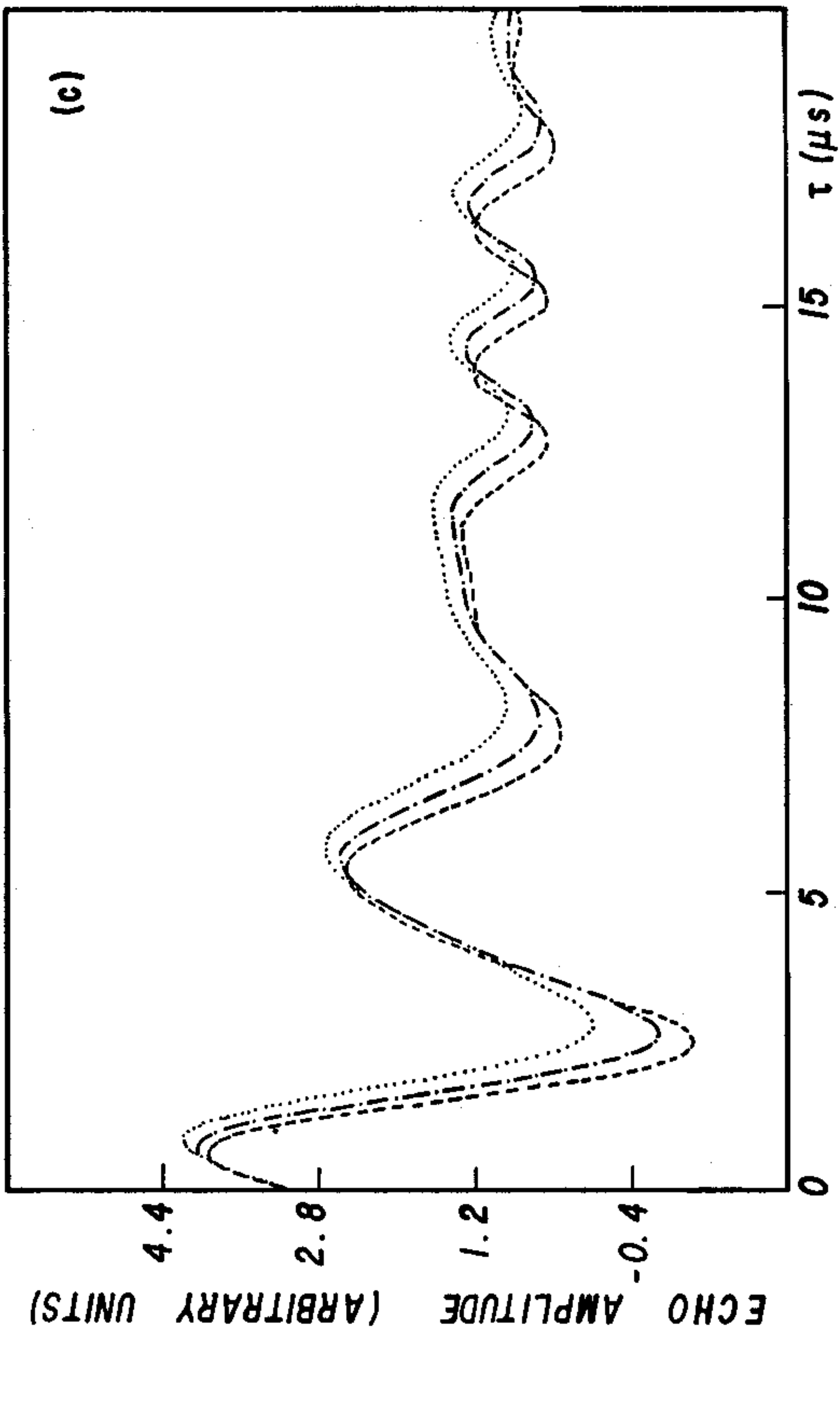

Fig. 8c

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