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RESPONSE OF EXCITON POLARITON SPECTRA AND ELECTRIC FIELDS TO DIFFERENT ADDITIONAL BOUNDARY CONDITIONS *

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ABSTRACT

The effects of three additional boundary conditions (ABC's) on the reflection and transmission spectra for exciton polaritons propagating in a spatially dispersive media are studied for both p and s configurations. An investigation of the ratios of the electric field amplitudes associated with the normal modes in these media is carried out. There is qualitative agreement among the predictions of the different ABC's, but there are significant quantitative differences, especially in the longitudinal polariton spike excited only in the p-geometry. Contact with formulations not using the ABC approach is made. The results are illustrated by parameters modelling the 1s exciton of PbI_2.

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I. INTRODUCTION

Spatial dispersion (SD) effects due to the wavevector dependence of the dielectric function have been extensively studied since the original phenomenological model of Pekar (1958) and Hopfield and Thomas (1963). The basic problem in media exhibiting SD is that more than one normal mode propagates in the crystal, thus giving rise to the so-called additional light waves (ALW). If the optical spectra are to be calculated, one needs to know the ratios of the electric field amplitudes of the ALW. There have been two approaches that have been used in such studies. One is to introduce the so-called additional boundary conditions (ABC's) in additions to Maxwell's boundary conditions (Pekar 1958, Zeyhar et al. 1972, Skettrup 1973, Biellmann 1974 amongst others). The other approach is to place emphasis on the non-local dielectric tensor and develop a formalism based on Maxwell's integro-differential equations (Agarwal et al. 1971a, 1971b, 1971c, Maradudin and Mills 1973, Bishop et al. 1976, Mukhopadhyay and Lundqvist 1977, Iadonisi et al. 1981 amongst others). The results of the ratios of the electric fields obtained in the second approach can be compared to those obtained by the ABC approach.

In this paper, we investigate the effects of three ABC's on reflection and transmission spectra and on the ratios of the ALW. The plan of the paper is as follows. In this introductory section we shall describe the geometry, the dipsersion relations, the electric fields associated with the ALW and the ABC's we are going to investigate. In Sec.II we shall discuss the reflection and transmission spectra for p-waves as well as the ratio functions. Sec.III will be devoted to a similar study for the s-geometry. Our results will be discussed in Sec.IV, taking PbI_2 as a model crystal.

A SD media can be described by a dielectric function $\epsilon(\omega,k)$ dependent on frequency ω and wavevector k and exciton mass M

$$\epsilon(\omega,k) = \epsilon_\infty + \frac{4\pi\beta\omega_0^2}{\omega_0^2 + \frac{\hbar\omega_0 k^2}{M} - \omega^2 - i\omega\Gamma} , \qquad (1)$$

where $4\pi\beta$ describes the strength of the resonance, ω_0 is the resonant frequency, Γ is the damping parameter. In all our illustrations we use $\Gamma = 0.1$ meV, which is below the critical damping for which SD effects are observable (Tait 1972, Nkoma 1980).

The geometry we are considering is illustrated in Fig.1, where in space $z > 0$ there is a non-dispersive media with a refractive index n_0, while in space $z < 0$ there is a crystal exhibiting SD effects. The ALW consists of transverse waves and a longitudinal mode. The transverse waves satisfy the dispersion relation:

$$\frac{c^2 k_\lambda^2}{\omega^2} = \frac{1}{2}[\epsilon_\infty - DE] \pm \frac{1}{2}\{[\epsilon_\infty + DE]^2 + 4EF\}^{1/2} \qquad (2)$$

where λ = 1,2 for p-waves or λ = 4,5 for s-waves, and the longitudinal mode (λ=3) satisfies

$$\frac{c^3 k^2_\lambda}{\omega^2} = (\omega^2 - \omega_3^2 + i\omega\Gamma)E \quad , \qquad (3)$$

where in (2) and (3) we have introduced

$$D = \omega_0^2 - \omega^2 - i\omega\Gamma \quad , \qquad (4a)$$

$$E = \frac{Mc^2}{\hbar\omega_0} \frac{1}{\omega^2} \quad . \qquad (4b)$$

$$F = 4\pi\beta\omega_0^2 \quad , \qquad (4c)$$

$$\omega_3^2 = \omega_0^2 + \frac{F}{\epsilon_\infty} \quad . \qquad (4d)$$

The dispersion curves are illustrated in Fig.2.

The electric fields associated with the ALW are of the form

$$E_\lambda \, e^{i(\underline{k}_\lambda \cdot \underline{r} - \omega t)} \qquad (5)$$

with the electric field amplitudes related by

$$E_2 = F_{21} E_1 \quad , \qquad (6a)$$

$$E_3 = F_{31} E_1 \quad , \qquad (6b)$$

$$E_5 = F_{54} E_4 \quad , \qquad (6c)$$

where the ratios F_{21}, F_{31} and F_{54} can be obtained by either of the approaches we have mentioned earlier. The wavevectors satisfy

$$k_\lambda^2 = k_{\lambda x}^2 + k_{\lambda z}^2 = \epsilon_\lambda \frac{\omega^2}{c^2} = n_\lambda^2 \frac{\omega^2}{c^2} \quad , \qquad (7a)$$

$$k_0^2 = k_{0x}^2 + k_{0z}^2 = n_0^2 \frac{\omega^2}{c^2} \quad , \qquad (7b)$$

where n_λ ia a refractive index, $k_{\lambda z}$ are normal components of wavevectors (not conserved), $k_{\lambda x}$ are tangential components of the wavevectors and are conserved, that is

$$k_{0x} = k_{1x} = k_{2x} = k_{3x} = k_{4x} = k_{5x} = n_0 \frac{\omega}{c} \sin\theta_0 \quad , \qquad (7c)$$

where θ_0 is the angle of incidence. Though the directions of the wavevectors in p and s configurations are different, their magnitudes are related according to

$$k_1 = k_4 \qquad (7d)$$

and

$$k_2 = k_5 \quad . \qquad (7e)$$

We shall investigate three most commonly used ABC's to be referred to as ABC1, ABC2 and ABC3. ABC1 is due to Pekar (1958) and requires that the total exciton polarization should vanish at the boundary

$$\sum_{\substack{\lambda = 1,2,3 \\ \text{or} \\ 4,5}} P_\lambda \Big|_{z=0} = 0 \quad . \qquad (8)$$

ABC2 (see, for example, Skettrup 1973) requires that the normal component of the derivative of the exciton polarization should vanish at the boundary

$$\sum_{\substack{\lambda = 1,2,3 \\ \text{or} \\ 4,5}} \frac{\partial P_\lambda}{\partial z}\Big|_{z=0} = 0 \qquad (9)$$

and ABC3 (see, for example, Biellmann et al. 1974) is a generalization of ABC1 and ABC2 in the form

$$\sum_{\substack{\lambda=1,2,3\\ \text{or}\\ 4,5}} \left[P_\lambda + B(\omega)\frac{\partial P_\lambda}{\partial z} \right]_{z=0} = 0 \quad , \tag{10}$$

where

$$B(\omega) = \frac{1}{i}\left\{\frac{\hbar\omega_0}{MD}\right\}^{1/2} \tag{11}$$

and D is given in (4a).

II. p-GEOMETRY SPECTRA

2.1 p-waves reflection spectra

By using Maxwell's boundary conditions (MBC) requiring the continuity of the tangential electric field component and normal displacement vector, the reflection coefficient can be written as

$$r_p = \frac{E_{rp}}{E_{0p}} = \frac{1 - n_p}{1 + n_p} \quad , \tag{12}$$

where the factor n_p contains ratios of ALW,

$$n_p = n_0^2 \frac{\left[\frac{f_1}{n_1} + \frac{f_2}{n_2} F_{21}^{(\alpha)} + \frac{1}{n_3} F_{31}^{(\alpha)}\right]}{\left[n_1 + n_2 F_{21}^{(\alpha)}\right]} \tag{13}$$

and

$$f_\lambda = \frac{k_{\lambda z}}{k_{0z}} \quad , \qquad \lambda = 1,2,3 \quad . \tag{14}$$

The expressions for $F_{21}^{(\alpha)}$ and $F_{31}^{(\alpha)}$ are for functions F_{21} and F_{31}, respectively, depending on whether ABC 1 (α=1), ABC 2 (α=2) or ABC 3 (α=3) has been used. The following results are obtained

$$F_{21}^{(1)} = -\frac{[\epsilon_1 - \epsilon_\infty][f_1 f_3 - \tan^2\theta_0]\, n_2}{[\epsilon_2 - \epsilon_\infty][f_2 f_3 - \tan^2\theta_0]\, n_1} \tag{15a}$$

$$F_{31}^{(1)} = \frac{n_3}{\epsilon_\infty}\left[(\epsilon_1 - \epsilon_\infty)\frac{f_1}{n_1} + F_{21}^{(1)}(\epsilon_2 - \epsilon_\infty)\frac{f_2}{n_2}\right] \tag{15b}$$

$$F_{21}^{(2)} = F_{21}^{(1)}\frac{f_1}{f_2} \tag{15c}$$

$$F_{31}^{(2)} = \frac{n_3}{\epsilon_\infty}\left[(\epsilon_1 - \epsilon_\infty)\frac{f_1^2}{n_1 f_3} + F_{21}^{(2)}(\epsilon_2 - \epsilon_\infty)\frac{f_2^2}{n_2 f_3}\right] \tag{15d}$$

$$F_{21}^{(3)} = F_{21}^{(1)}\frac{[1 + i\,B(\omega)\,k_{1z}]}{[1 + i\,B(\omega)\,k_{2z}]} \tag{15e}$$

$$F_{31}^{(3)} = \frac{n_3}{\epsilon_\infty}\left[(\epsilon_1 - \epsilon_\infty)\frac{[1 + i\,B(\omega)\,k_{1z}]f_1}{[1 + i\,B(\omega)\,k_{3z}]n_1} + F_{21}^{(3)}(\epsilon_2 - \epsilon_\infty)\frac{[1 + i\,B(\omega)\,k_{1z}]f_2}{[1 + i\,B(\omega)k_{3z}]n_1}\right] \tag{15f}$$

Fig.3 illustrates the p-waves reflection spectra as calculated using ABC 1, ABC 2 and ABC 3 functions at angles of incidence of 30° and 45°. The ratio functions $F_{21}^{(\alpha)}$ and $F_{31}^{(\alpha)}$ for α = 1,2,3 are given in (15a) to (f) and their magnitudes are illustrated graphically in Figs.4(a), (b). A further discussion will follow in Sec.IV.

2.2 p-waves transmission spectra

Since there are three ALW in the p-geometry, three transmission coefficients have to be calculated for a SD medium; t_1 and t_2 for modes 1 and 2 which are transverse and t_3 for the longitudinal mode 3. By using MBC one obtains

$$t_1 = \frac{E_1}{E_{0p}} = \frac{2}{\left[\frac{n_1}{n_0} + f_1\frac{n_0}{n_1}\right] + \left[\frac{n_2}{n_0} + f_2\frac{n_0}{n_2}\right]F_{21}^{(\alpha)} + \frac{n_0}{n_3}F_{31}^{(\alpha)}} \tag{16}$$

$$t_2 = \frac{E_2}{E_{0p}} = t_1 F_{21}^{(\alpha)} \qquad (17)$$

$$t_3 = \frac{E_3}{E_{0p}} = - t_1 F_{31}^{(\alpha)} \text{Cotan}\ \theta_0 \ , \qquad (18)$$

The dependence of the transmission spectra on the three ABC's is shown in Figs.5(a), (b) and (c) and it can be seen that they agree qualitatively, but differ quantitatively. In all cases t_1 decreases as frequency increases, t_2 has two close peaks around ω_3. The longitudinal mode spectra (t_3) is shown in Fig.6, where it can be seen that the ABC 2 peak is much lower than the peaks due to ABC 1 and ABC 3.

III. s-WAVES SPECTRA

3.1 s-waves reflection spectra

Application of MBC results to a reflection coefficient of the form

$$r_s = \frac{E_{rs}}{E_{0s}} = \frac{1 - n_s}{1 + n_s} \ , \qquad (19)$$

where n_s contains ratios of mode 4 and mode 5,

$$n_s = \frac{f_4 + F_{54}^{(\alpha)} f_5}{1 + F_{54}^{(\alpha)}} \ , \qquad (20)$$

where f_4 and f_5 are given by (14) for $\lambda = 4,5$. The ratio $F_{54}^{(\alpha)}$ is dependent on the ABC used, and the results are

$$F_{54}^{(1)} = - \frac{[\epsilon_4 - \epsilon_\infty]}{[\epsilon_5 - \epsilon_\infty]} \qquad (21a)$$

$$F_{54}^{(2)} = F_{54}^{(1)} \frac{f_4}{f_5} \qquad (21b)$$

$$F_{54}^{(3)} = F_{54}^{(1)} \frac{[1 + i B(\omega) k_{4z}]}{[1 + i B(\omega) k_{5z}]} \ . \qquad (21c)$$

Fig.7 illustrates the s-waves reflection spectra based on the three ABC's. The frequency dependence of the magnitude of the ratio functions 21(a),(b) and (c) is illustrated in Fig.8. We shall discuss these aspects in more detail in Sec.IV.

3.2 s-waves transmission spectra

Two transverse modes are excited in this geometry and no longitudinal mode is excited. Applying MBC, the transmission coefficients corresponding to modes 4 and 5 are given by t_4 and t_5 in the form

$$t_4 = \frac{E_4}{E_{0s}} = \frac{2\,(f_5 - n_s)}{(f_5 - f_4)(1 + n_s)} \qquad (22)$$

$$t_5 = \frac{E_5}{E_{0s}} = \frac{2\,(f_4 - n_s)}{(f_4 - f_5)(1 + n_s)} \ , \qquad (23)$$

where n_s has been given by (20) and f_4, f_5 are of the form (14). Fig.9 illustrates the frequency dependence of $|t_4|$ and $|t_5|$ on the three ABC's. The s-waves transmission spectra differ from that of the p-waves spectra since in the s-geometry the longitudinal mode is not excited.

IV. DISCUSSION

In this paper we have investigated the sensitivity of reflection and transmission spectra to ABC's (8), (9) and (10) for both the p-geometry and s-geometry. Our results are illustrated by using the parameters modelling the Is exciton of PbI_2 as given by Tosatti and Harbeke (1974); $\omega_0 = 2.497$ eV, $\epsilon_\infty = 12.0$, $4\pi\beta = 0.065$ and $M = 1.5\, m_0$. It is observed that the spectra are dependent on the ABC's used through the factors $F_{21}^{(\alpha)}$, $F_{31}^{(\alpha)}$ and $F_{54}^{(\alpha)}$. The spectra agree qualitatively but differ quantitatively, and in particular the peaks predicted by ABC 2 are always higher than those of the other ABC's as can be seen in Figs.3,5,6,7, and 9. Another recurring feature is that ABC 3 spectra always cross ABC 1 spectra at a frequency below ω_0. Generally all the ABC's have a closer agreement at frequencies $\omega >> \omega_0$.

In Figs.4 and 8 we have plotted the frequency dependency of the magnitudes of the ratio functions $F_{21}^{(\alpha)}$, $F_{31}^{(\alpha)}$ and $F_{54}^{(\alpha)}$ for frequencies above ω_3 when all the modes propagate simultaneously. It can be concluded that at such frequencies mode 2 is most intense followed by mode 3 and mode 1 is least intense for the p-configuration, while in the s-configuration mode 5 is more intense than mode 4. However, the magnitudes differ significantly since $F_{21}^{(2)}$ differs from $F_{21}^{(1)}$, $F_{21}^{(3)}$ by a factor of ten, while F_{31} values are close for all the three ABC's. This can be understood from Eq.(15a) to Eq.(15f), where for example it can be concluded that

$$F_{21}^{(2)} > F_{21}^{(1)} \text{ always, since } f_1 > f_2 \qquad (24)$$

as can be deduced from Eqs.(7a,b), (15a,c) and the dispersion curves in Fig.2.

It is in order to comment on the problem of the ABC approach viz the alternative approach based on Maxwell's integro-differential equations (MIDE) The ABC's are known to be introduced in a rather ad hoc basis and there have always been attempts to find some microscopic justification of the process. In the MIDE approach this is thought to be unnecessary. However, one can note that the main results for the reflection and transmission spectra remain the same except for the change

$$F_{\beta 1}^{(\alpha)} \longrightarrow F_{\beta 1}^{\text{other}} \quad ; \quad \beta = 2,3 \qquad (25a)$$

$$F_{54}^{(\alpha)} \longrightarrow F_{54}^{\text{other}} \qquad (25b)$$

in Eq.(13) for p-waves and Eq.(20) for s-waves. In Eqs.(25a,b), $F_{\beta 1}^{\text{other}}$ or F_{54}^{other} would be the new functions to be calculated within the MIDE formalism, or indeed any other formalism. If the alternative approaches do not produce similar factors, then a similar study carried here can be done.

The factors $F_{\beta 1}^{(\alpha)}$ and $F_{54}^{(\alpha)}$ also affect other optical spectra, such as Brillouin scattering spectra (Yu 1979, Tilley 1980), and the points made in (25a,b) are equally valid.

Finally it should be emphasized that exciton polariton spectra are very sensitive to damping and SD effects are sensitive only below a certain critical damping (Tait 1972). In this paper, the value of damping is below such a critical damping for PbI_2 as discussed elsewhere (Nkoma 1980) in connection with absorption by exciton polaritons.

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FIGURE CAPTIONS

Fig.1 E_{0p} (E_{0s}) and E_{rp} (E_{rs}) are incident and reflected electric fields, respectively, for the p-geometry (s-geometry). Modes 1,2,3 are excited in the p-geometry while modes 4,5 are excited in the s-geometry.

Fig.2 Dispersion curves for exciton polaritons in PbI_2 showing dispersion of modes 1,2,3,4 and 5.

Fig.3 Reflection spectra for PbI_2 calculated using ABC 1 (⊖⊖⊖) ABC 2 (•—•—•) and ABC 3 (✕✕✕) at $\theta_0 = 30^\circ, 45^\circ$ for the p-geometry. The arrow (↓) in this and other figures shows the position of the longitudinal frequency.

Fig.4(a) Frequency dependence of the magnitude of the ratio functions, $|F_{21}^{(\alpha)}|$ and $|F_{31}^{(\alpha)}|$ for PbI_2 calculated using ABC 1 (⊖⊖⊖), ABC 2 (•—•—•) and AMC 3 (✕✕✕) at $\theta_0 = 30^\circ$ for p-geometry.

Fig.4(b) Same caption as Fig.4(b) except that $\theta_0 = 45^\circ$.

Fig.5(a) Transmission spectra for transverse modes 1 and 2 in PbI_2 calculated using ABC 1 at $\theta = 30^\circ$ in p-geometry.

Fig.5(b) Transmission spectra for transverse modes 1 and 2 in PbI_2 calculated using ABC 3 at $\theta = 30^\circ$ in p-geometry.

Fig.5(c) Transmission spectra for transverse modes 1 and 2 in PbI_2 calculated using ABC 2 at $\theta = 30^\circ$ in p-geometry.

Fig.6 Transmission spectra for longitudinal modes 3 in PbI_2 using ABC 1 (⊖⊖⊖), ABC 2 (•—•—•) and ABC 3 (✕✕✕) at $\theta_0 = 30^\circ, 45^\circ$ in p-geometry.

Fig.7 Reflection spectra for PbI_2 calculated using ABC 1 (⊖⊖⊖), ABC 2 (•—•—•) and ABC 3 (✕✕✕) at $\theta_0 = 30^\circ, 45^\circ$ for the s-geometry.

Fig.8 Frequency dependence of the magnitudes of the ratio function $|F_{54}^{(\alpha)}|$ using ABC 1 (⊖⊖⊖), ABC 2 (•—•—•) and ABC 3 (✕✕✕) at $\theta_0 = 30^\circ, 45^\circ$ for the s-geometry.

Fig.9 Transmission spectra for transverse modes 4 and 5 in PbI_2 calculated using ABC 1 (⊖⊖⊖), ABC 2 (•—•—•) and ABC 3 (✕✕✕) at $\theta_0 = 30^\circ, 45^\circ$ for the s-geometry.

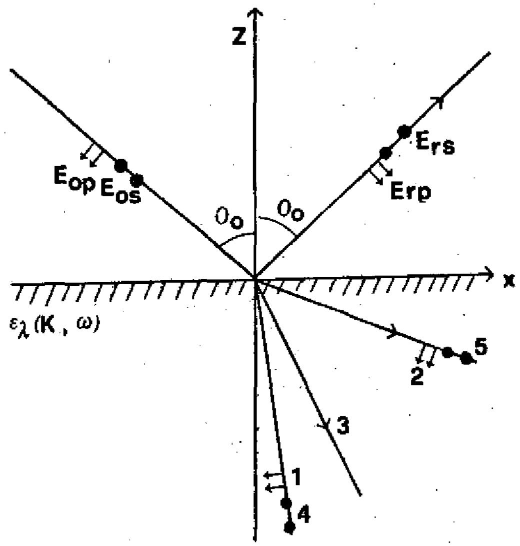

Z
x
E_{op} E_{os}
E_{rs}
E_{rp}
0_o
0_o
$\varepsilon_\lambda(K, \omega)$
1
2
3
4
5

Fig.1

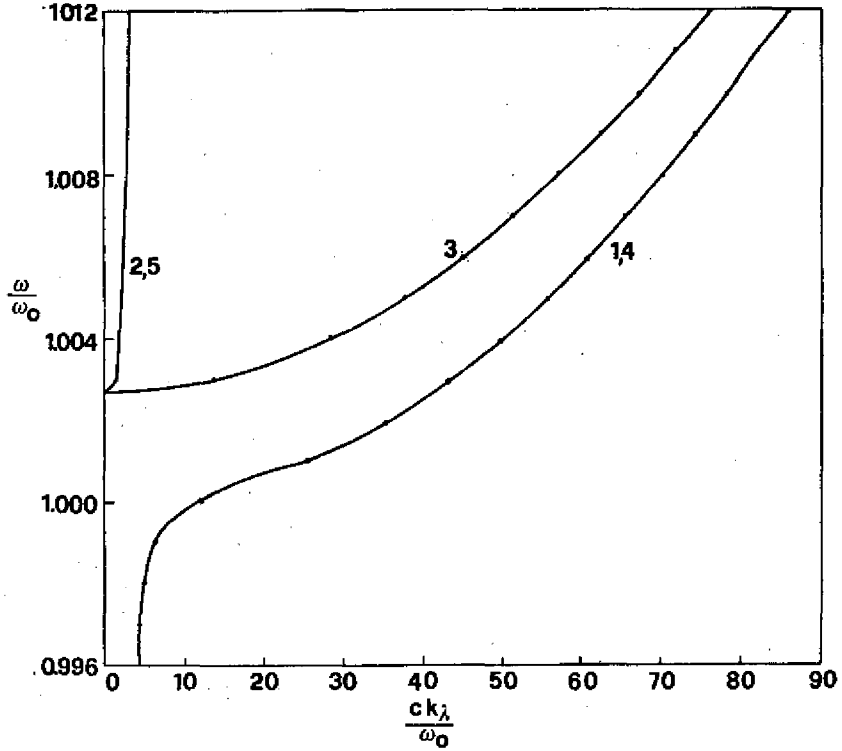

1.012
1.008
1.004
1.000
0.996
$\frac{\omega}{\omega_o}$
0
10
20
30
40
50
60
70
80
90
$\frac{ck_\lambda}{\omega_o}$
2,5
3
1,4

Fig.2

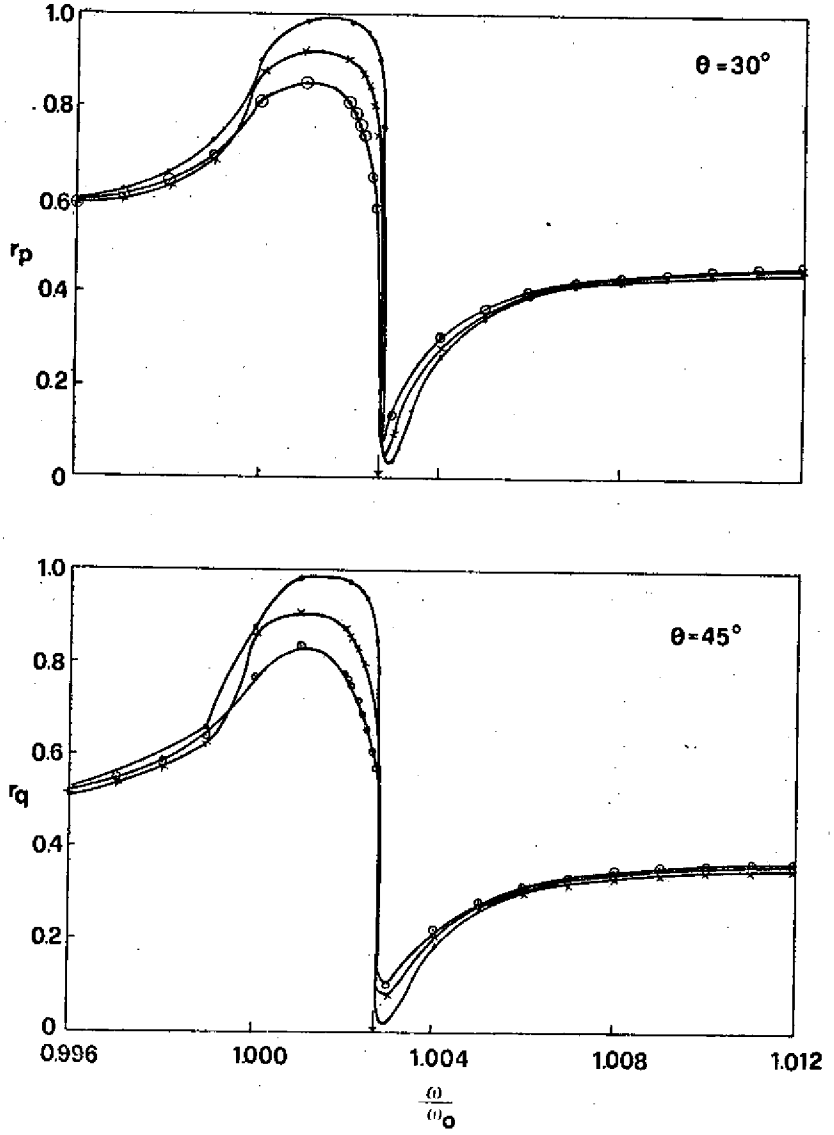

θ=30°
θ=45°
r_p
r_q
1.0
0.8
0.6
0.4
0.2
0
0.996
1.000
1.004
1.008
1.012
$\frac{\omega}{\omega_o}$

Fig.3

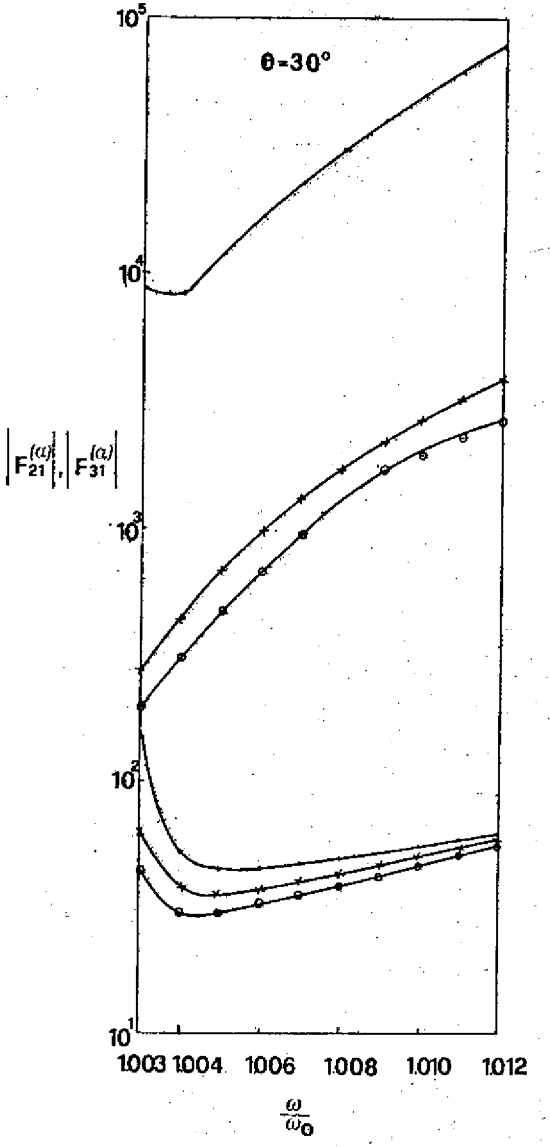
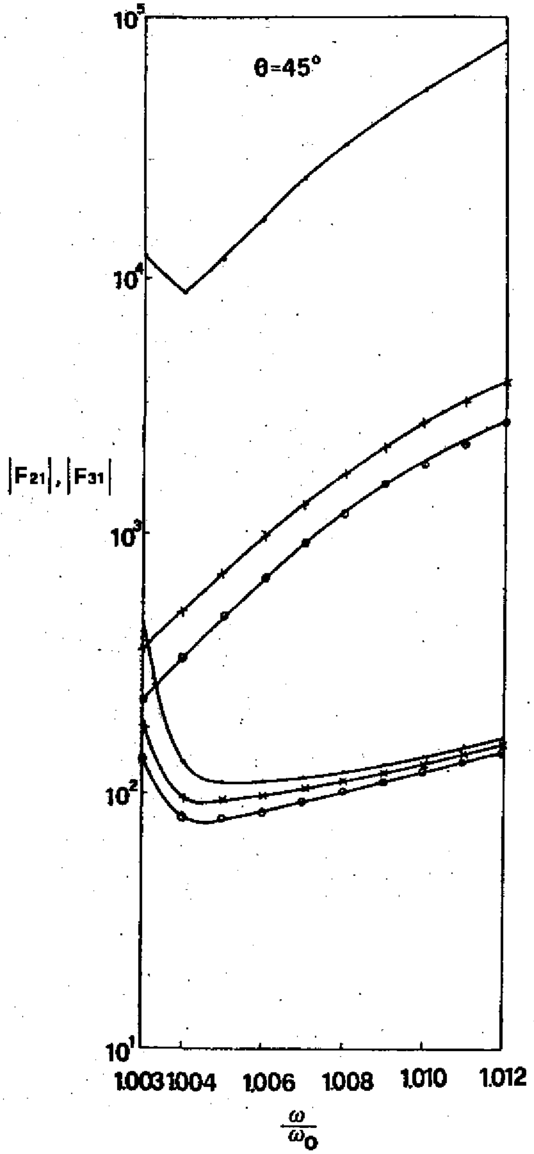

θ=30°
10^5
10^4
10^3
10^2
10^1
$\left|F_{21}^{(\alpha)}\right|, \left|F_{31}^{(\alpha)}\right|$
1.003
1.004
1.006
1.008
1.010
1.012
$\frac{\omega}{\omega_o}$

Fig4 a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10^4
10^3
10^2
10^1
|F21|, |F31|
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1.004
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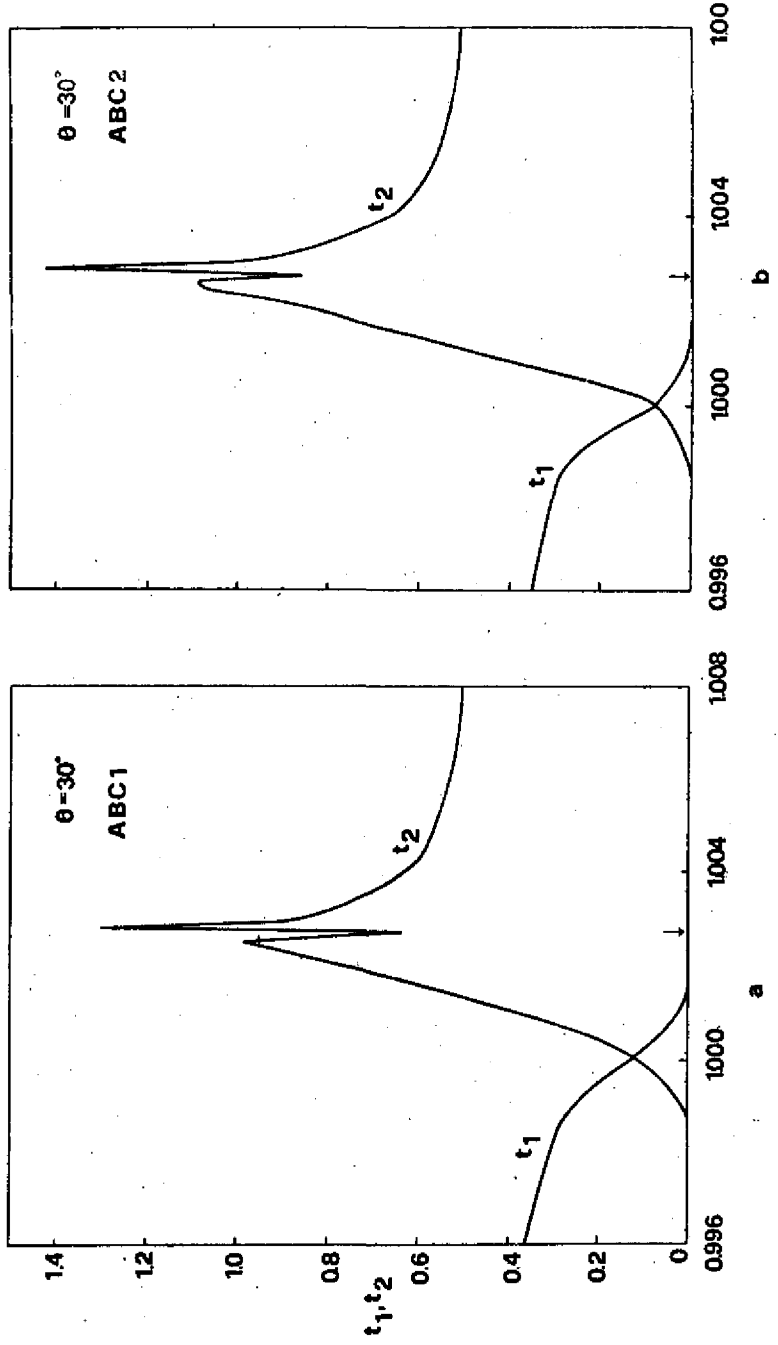
1.008
1.010
1.012
$\frac{\omega}{\omega_0}$

Fig4b


θ=30°
ABC 2
θ=30°
ABC 1
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t_1
t_2
t_1
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1.2
1.0
0.8
0.6
0.4
0.2
0
t_1, t_2
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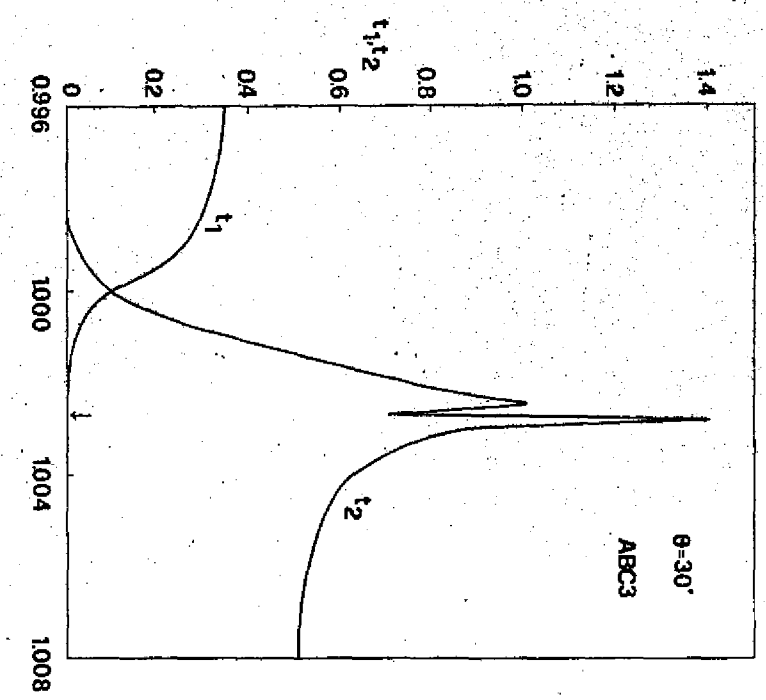
1.000
1.004
1.008
a
b

Fig 5



t₁,t₂
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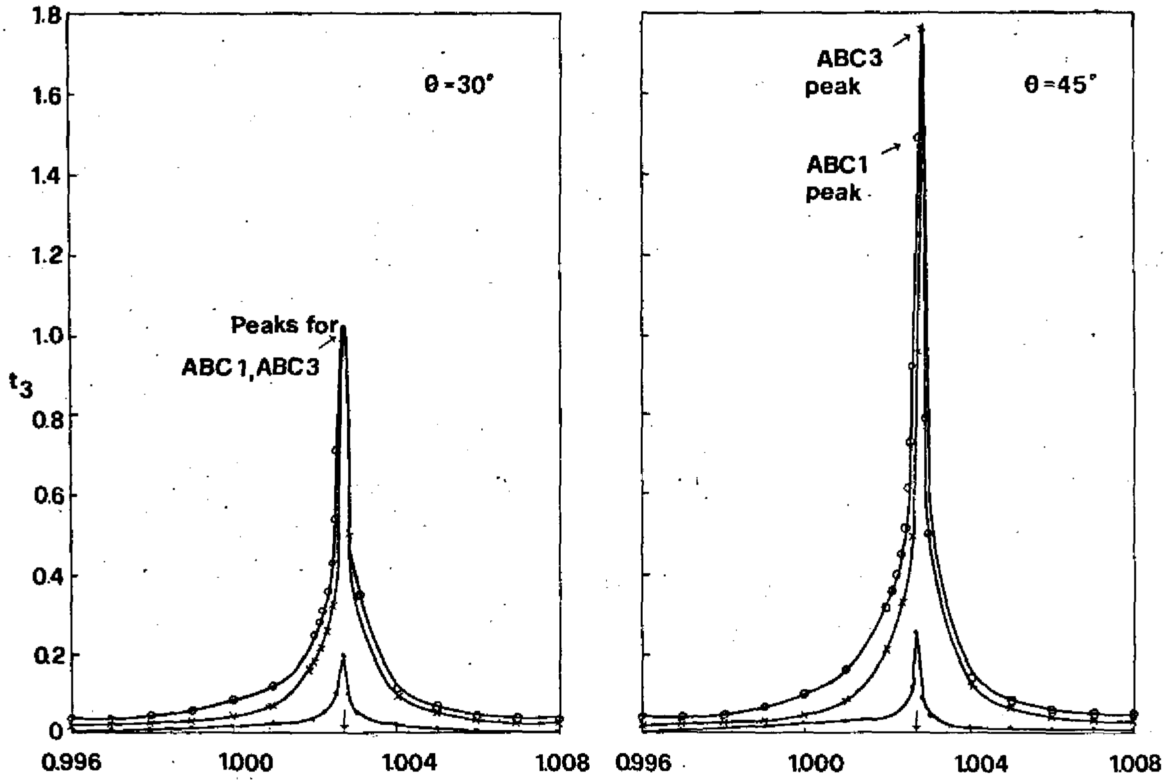
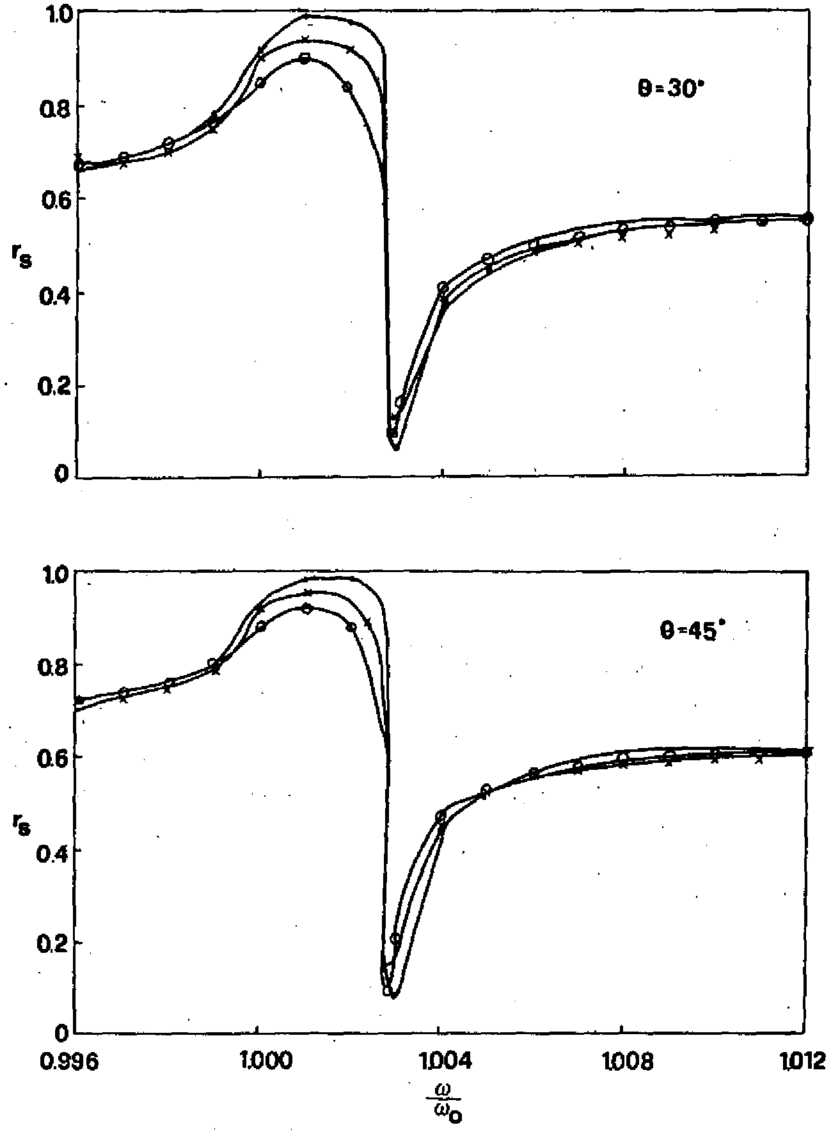
0.2
0.4
0.6
0.8
1.0
1.2
1.4
0.996
1.000
1.004
1.008
t₁
t₂
θ=30°
ABC3

Fig.5c



t₃
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
0.996
1.000
1.004
1.008
θ=30°
Peaks for
ABC1,ABC3
0.996
1.000
1.004
1.008
θ=45°
ABC3
peak
ABC1
peak

Fig.6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0.6
0.4
0.2
0
r_s
θ=30°
θ=45°
0.996
1.000
1.004
1.008
1.012
$\frac{\omega}{\omega_0}$

Fig.7

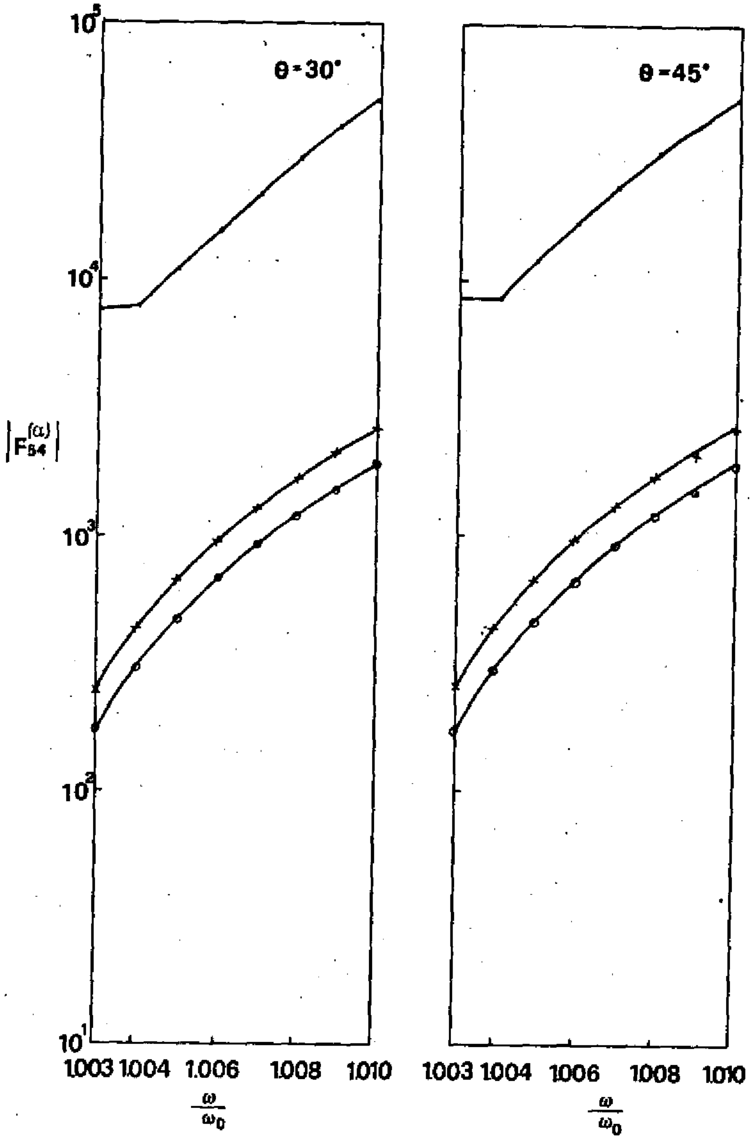
10^5
10^4
10^3
10^2
10^1
$\left|F_{54}^{(\alpha)}\right|$
θ=30°
θ=45°
1.003
1.004
1.006
1.008
1.010
1.003
1.004
1.006
1.008
1.010
$\frac{\omega}{\omega_0}$
$\frac{\omega}{\omega_0}$

Fig.8

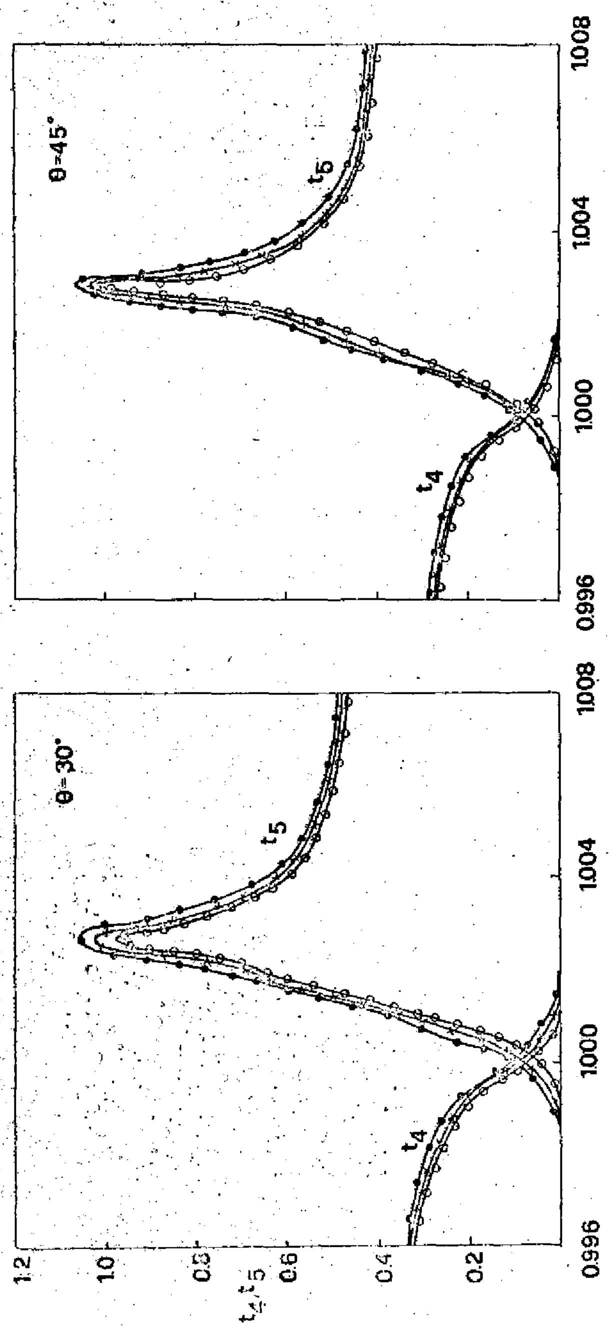

Fig. 9

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