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Electron-Phonon Interaction and Raman Scattering in High T_c Oxides

Debanand Sa[1]

Institute of Physics, Bhubaneswar-751005, INDIA

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Abstract

A new kind of electron-phonon interaction for the high T_c oxides besides the usual one is proposed. This work which is motivated by the recent Raman data for $YBCO$, supports the local pair model of superconductivity. The spectral density function for both the phonons are calculated in the normal state as well as in the superconducting state. Finally, the Raman intensity is computed and the temperature dependence of the phonon frequency shift and the linewidth is compared in a qualitative way with the experimental results.

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[1]e-mail:debanand@iopb.ernet.in

Raman scattering in high T_c cuprates [1] shows several unusual features in the normal state as well as in the superconducting state. In the normal state, a rather large but constant background intensity has been observed at all frequencies up to the highest measured one, which has been attributed to the scattering of the charge carriers in the system [2]. Three prominent phonon peaks having frequencies of $116cm^{-1}$, $340cm^{-1}$ and $504cm^{-1}$ ride over the constant background intensity out of which some show Fano line shape indicating a strong electron-phonon interaction in the system. On lowering the temperature below the superconducting transition temperature (T_c), the constant background intensity is suppressed at low frequencies which may be a signature of an appearence of a superconducting gap in the system. Moreover, some of the phonons show anomalous softening but other hardens. It seems that the temperature dependence of the Raman active modes in the layered cuprates are very much sensitive to the oxygen concentration (δ). Recently Altendorf et al [3] have studied the temperature dependence of the $340cm^{-1}$ and $440cm^{-1}$, $500cm^{-1}$ phonons in different crystals of $YBa_2Cu_3O_\delta$ ($6.7 \leq \delta \leq 7.0$). From the measurement of the Raman shift, the linewidth and the Fano asymmetry parameter, they observed that the above two sets of phonons show almost opposite behaviour to each other which of course, strongly depend on the doping concentration and hence it is argued that the $440cm^{-1}$ and $500cm^{-1}$ phonons interact with a different set of electrons than does the $340cm^{-1}$ phonon.

A theoretical model for the normal state as well as the superconducting state is needed for an explanation of the Raman data. Unfortunately, at present time we are far

away from the proper understanding of the mechanism of high T_c superconductivity even though many theoretical models have been proposed [4]. However, due to the anomalous normal state properties it is believed that the usual BCS formalism may not be a good candidate to explain these materials. The observation of small coherence length indicates that the pairing of the charge carriers must be reasonably localised in the coordinate space [5]. Of course, the possibilities for the scenario of superconductivity in the layered cuprates based on the local pair rather than the Cooper pair has already been discussed [6-14]. The normal state of the local pair superconductors is characterised by an interaction of the local bosons with the conduction electrons. Superconductivity appears here due to the bose condensation of the local pairs which may also induce Cooper pairing. The remarkable feature of the local pair superconductors is that it yields the anomalous polarisability in the normal state [15], similar to that of the phenomenological marginal Fermi liquid hypothesised by Varma et al [2].

In order to calculate the Raman scattering amplitude, it is necessary to evaluate the phonon response in the system which will depend on the coupling of phonons to the charge carriers. Being motivated by the recent Raman data for $YBCO$ [3] which of course favours the local pair model of superconductivity, a new kind of electron-phonon interaction besides the usual one is proposed. In the usual case, phonon couples to the charge carrier density whereas in the present one it interacts with the charge carrier pairs in the normal state. Thus it is expected to modify the phonon spectrum in different ways since the self energy in both the cases will differ remarkably. The Hamiltonian for

such a coupled electron-phonon system in an ad hoc basis is given as

$$H = H_o + H_I^1 + H_I^2 \tag{1}$$

where

$$H_o = \sum_{k,\sigma} \epsilon_k c^\dagger_{k,\sigma} c_{k,\sigma} + \sum_q \omega_q a_q^{1\dagger} a_q^1 + \sum_q \Omega_q a_q^{2\dagger} a_q^2 \tag{2}$$

$$H_I^1 = g_1 \sum_{k,q,\sigma} c^\dagger_{k+q,\sigma} c_{k,\sigma} (a_q^1 + a_{-q}^{1\dagger}) \tag{3}$$

$$H_I^2 = g_2 \sum_{k,q,\sigma} (c^\dagger_{k-q,\sigma} c^\dagger_{-k,-\sigma} + c_{-k-q,-\sigma} c_{k,\sigma})(a_q^2 + a_{-q}^{2\dagger}) \tag{4}$$

$c^\dagger_{k,\sigma}(c_{k,\sigma})$ is the creation(annihilation) operator for an electron with momentum $\vec{k}$ and spin σ, ϵ_k is the single particle energy; $a_{-q}^{1,2\dagger}(a_q^{1,2})$ are the qth creation (annihilation) operators for phonon 1,2 with the frequencies ω_q and Ω_q respectively. Phonon 1 is the propagating one as in the usual metals, g_1 being its coupling strength whereas phonon 2, we call it as local since it couples to the charge carrier pairs and its coupling is g_2. The derivation of such a kind of interaction Hamiltonian for phonons with the charge carriers in a correlated metallic state and the modification of the phonon spectrum is discussed in an earlier paper [16].

Due to the appearence of a normal state different from that of the Fermi liquid like metals as well as the interaction of local phonon with the charge carrier pairs, the usual phonon-Raman scattering process needs to be modified. Usually, the scattering of light [17] in normal metals proceeds as follows: The incident photon excites an electron-hole pair, which either emits or absorbs a phonon due to the electron-phonon interaction in the system. Finally, the electron and hole recombine giving rise to the scattered photons.

Therefore, in such a process the scattered intensity of the photon will be proportional to the sum of the imaginary part of the particle-hole polarisability and the phonon spectral density function. But in the present case the scenario is different due to the appearence of a different normal state which is characterised by local pairs. Therefore it makes the situation a bit more complicated. The incident photon besides exciting the particle-hole pairs, can also break the local pairs which gives rise to an anomalous polarisability in the particle- particle channel. Moreover, this process will also couple to phonons as seen from eqn.(4). Thus in such a normal state, the Raman intensity will acquire an additional contribution coming from the sum of the imaginary part of the pair polarisability and the spectral density function for the local phonons. The expression for the Raman intensity $I(\omega)$ therefore becomes

$$I(\omega) \sim (1+n(\omega))[-Im\chi^{p-h}(0,\omega) - Im\chi^{p-p}(0,\omega) + S^1(0,\omega) + S^2(0,\omega)] \qquad (5)$$

where $n(\omega)$ is the Bose factor, $\chi^{p-h,p-p}(0,\omega)$ being the normal state polarisabilities in the particle-hole and the particle-particle channel respectively and $S^{1,2}(0,\omega)$ are the corresponding spectral density functions for the propagating and the local phonons. In general, the spectral density function is related to the imaginary part of the phonon Green's function $D(0,\omega)$ as

$$S(0,\omega) = -2ImD(0,\omega). \qquad (6)$$

Both the phonon Green's functions which involve the calculation of the phonon self

energies can be evaluated with the help of the diagonal part of the Hamiltonian and the spectral density function is calculated. With the knowledge of the polarisabilities and the phonon spectral density functions in the normal state as well as the superconducting state, Raman intensity is computed in the low and high temperature limits.

The basic feature observed in the Raman intensity from Figs.1-2 is the appearence of a constant background intensity in the normal state as well as the superconducting state, which can be attributed to the scattering of charge carriers in the system since it is an outcome of the polarisability in the particle-particle channel [2, 19]. Moreover, the propagating as well as the local phonons ride over the background intensity. As it can be seen from Figs.1(a,b) which is the Raman intensity of the normal state for the case $\omega < T$ that there are changes in the frequency as well as in the linewidth of the local phonon on varying the temperature. There are almost no changes for the case of the propagating phonon in the normal state (the small changes seen in Fig.1 may be due to the interference of the local phonon) since its self energy in the lowest order vanishes for $q \to 0$. Of course, for convenience a phenomenological width (η_1, η_2) has been provided to both the phonons in the normal state as well as in the superconducting state besides the natural one which comes out from the imaginary part of the phonon self energies. On reducing the temperature in the normal state, the intensity of the local phonon decreases and its frequency hardens with broadening in the linewidth for the frequency of the local phonon equals the twice of the propagating one $(\Omega_o = 2\omega_o)$(Fig.1(a)) but in the case where the local phonon frequency is half of the propagating one $(\omega_o = 2\Omega_o)$(Fig.1(b)) it

softens with broadening in the linewidth. The softening which is in accordance with the experimental observed spectrum [3] could be associated with the local lattice distortion due to the presence of the local pairs in the normal state. Of course, an appearence of a normal state gap in the spectrum can also provide an account for the softening above T_c[18]. Moreover, the broadening in the linewidth is expected due to the pair breaking in the normal state since the temperature here will be higher than the superconducting gap energy ($T > 2\Delta$).

In general, the temperature dependence of the frequencies and the linewidths of the Raman active modes can be accomplished by determining the phonon self energy changes that occur below the critical temperature. The imaginary part of the self energy is proportional to the phonon linewidth (or inverse lifetime) which is measured as a function of temperature. If the phonon energy is less than the superconducting gap energy ($\Omega_o, \omega_o < 2\Delta$), it can not decay by pair breaking since there will be a decrease in the number of available decay channels and thus a decrease in the linewidth. On the otherhand, if the phonon energy is greater than the gap energy ($\Omega_o, \omega_o > 2\Delta$), it will be having sufficient energy to dissociate an electron/hole pair and the number of possible decay channels can be increased, resulting in an increase in the linewidth. In a similar way, the real part of the self energy is proportional to the phonon energy and the superconductivity induced changes are determined from a measurement of the phonon frequency as a function of the temperature below T_c. Qualitatively for phonon energy below or near the gap ($\Omega_o, \omega_o < 2\Delta$), the electron-phonon interaction pushes the

frequencies to smaller values which causes softening. On the contrary, for $\Omega_o, \omega_o > 2\Delta$, the frequency is expected to increase and hence it hardens.

In the present case, it is observed that on reducing the temperature in the superconducting state, the frequency of the local phonon softens for $\omega_o < \Omega_o < 2\Delta$ (Fig.2(a)), whereas for $\Omega_o > \omega_o > 2\Delta$ (Fig.2(b)), it hardens without any softening/hardening in the propagating phonon. Moreover, the linewidth of the local as well as the propagating phonons in both the cases broadens which accounts in favour of the heavily doped samples of $YBCO$ by Altendorf et al [3]. On the otherhand, in the case where $\omega_o < 2\Delta < \Omega_o$ (Fig.2(c)), both the phonons soften with broadening in the linewidth but for $\Omega_o < 2\Delta < \omega_o$ (Fig.2(d)), the frequency of the local phonon softens and that of the propagating one hardens with linewidth broadening, resulting in qualitative agreement with the experimentally observed spectrum [3]. In fact, the role of the lattice anharmonicities can not be ruled out completely for the explanation of the unusual features seen in the Raman spectrum in high T_c oxides. Moreover, it is observed from Figs.1-2 that the spectrum shows asymmetry line shape for both the phonons which is indicative of a strong electron-phonon interaction in the system. This is in favour of the interaction of the local phonon with the charge carrier pairs as observed in Figs.1 and 2 whose coupling is stronger than that of the propagating one.

Recent Raman measurements on the frequency shift and the linewidth in the high T_c oxides have been compared with the result of the strong coupling theory of Zeyher and Zwicknagl(ZZ) [20]. It is estimated that the superconducting gap which is relatively

large, increases from about $2\Delta = 5.4k_BT_c$ to $2\Delta = 7.0k_BT_c$ for decreasing the oxygen concentration from 7.0 to 6.9 [3]. In contrast, the present work which is rather qualitative not quantitative, therefore the estimation of the superconducting gap here is not feasible. Moreover, it differs from that of ZZ on the fact that the normal state here is not the usual Fermi liquid as well as the presence of the two different kinds of electron-phonon interactions modify the phonon spectrum differently, which qualitatively agrees with the experimentally observed spectrum. But to quantify it, one has to include the doping concentration as an explicit parameter in the theory to carry out the Raman intensity, since the temperature dependence of the frequency shift and the linewidth of the $340cm^{-1}$,$440cm^{-1}$ and $500cm^{-1}$ phonons is quite sensitive to the doping concentration.

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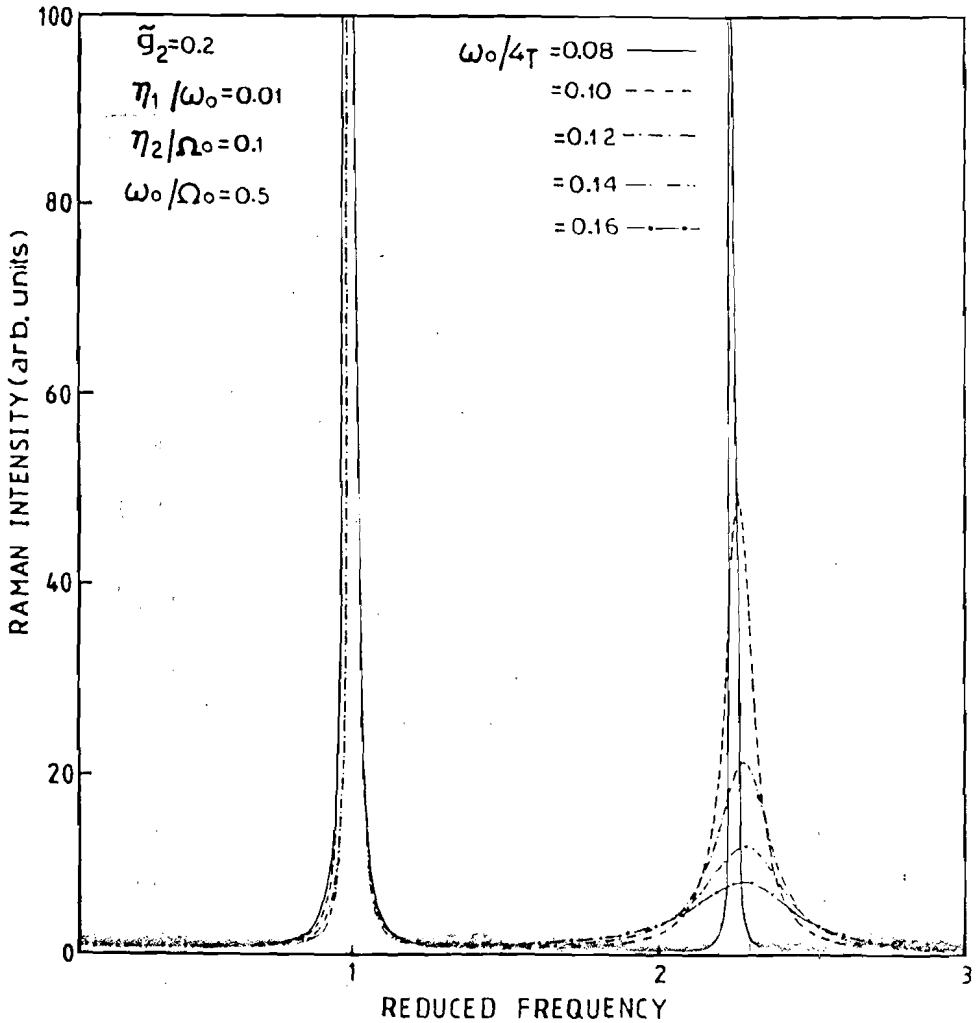
FIGURE CAPTIONS

Figure 1: Raman Intensity in the normal state as a function of the reduced frequency: (a) $\Omega_o = 2\omega_o$ and (b) $\omega_o = 2\Omega_o$.

Figure 2: Raman Intensity in the superconducting state as a function of the reduced frequency: (a) $\omega_o < \Omega_o < 2\Delta$, (b) $\Omega_o > \omega_o > 2\Delta$, (c) $\omega_o < 2\Delta < \Omega_o$ and (d) $\omega_o > 2\Delta > \Omega_o$.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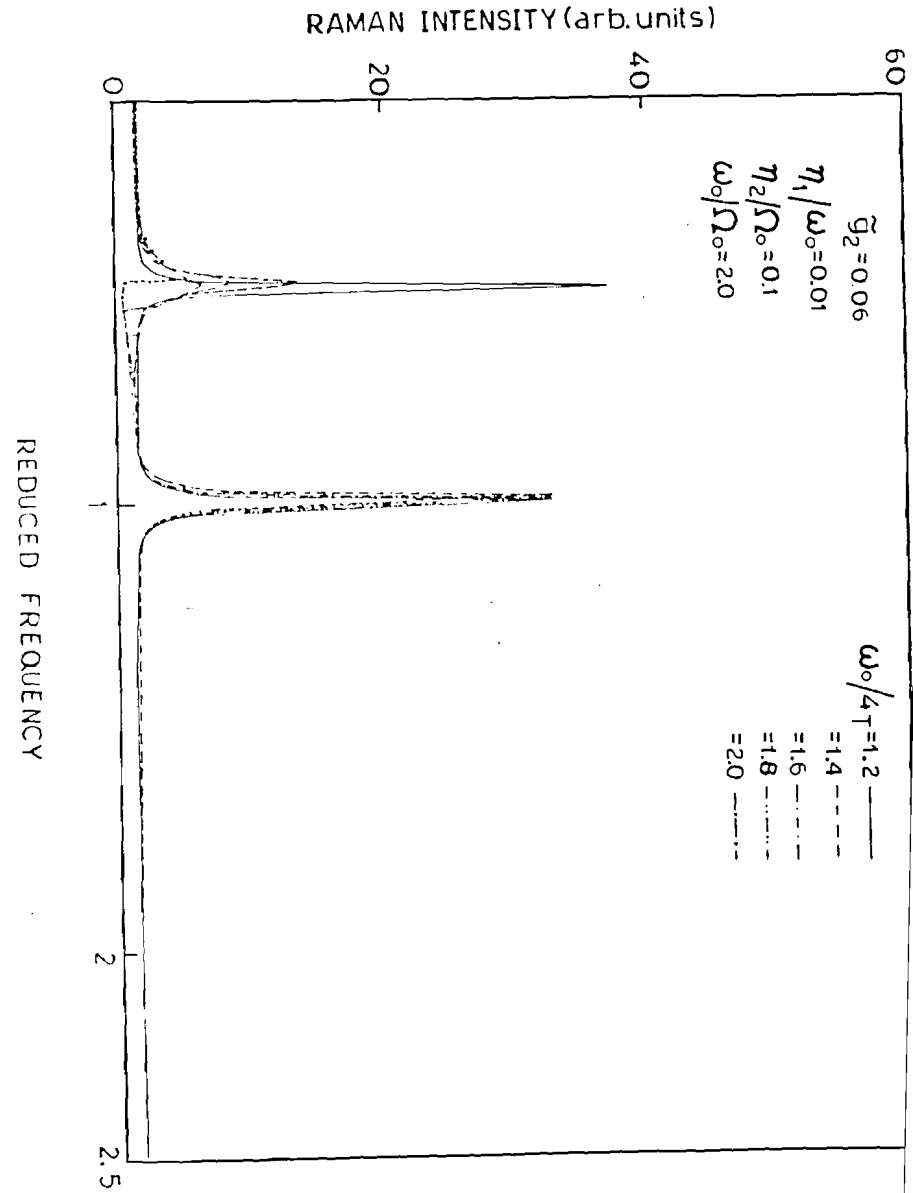
η_2/Ω_0=0.1
ω_0/Ω_0=0.5
$\omega_0/4_T$ =0.08
=0.10
=0.12
=0.14
=0.16
RAMAN INTENSITY (arb. units)
REDUCED FREQUENCY
0
20
40
60
80
100
1
2
3

Fig.1(a)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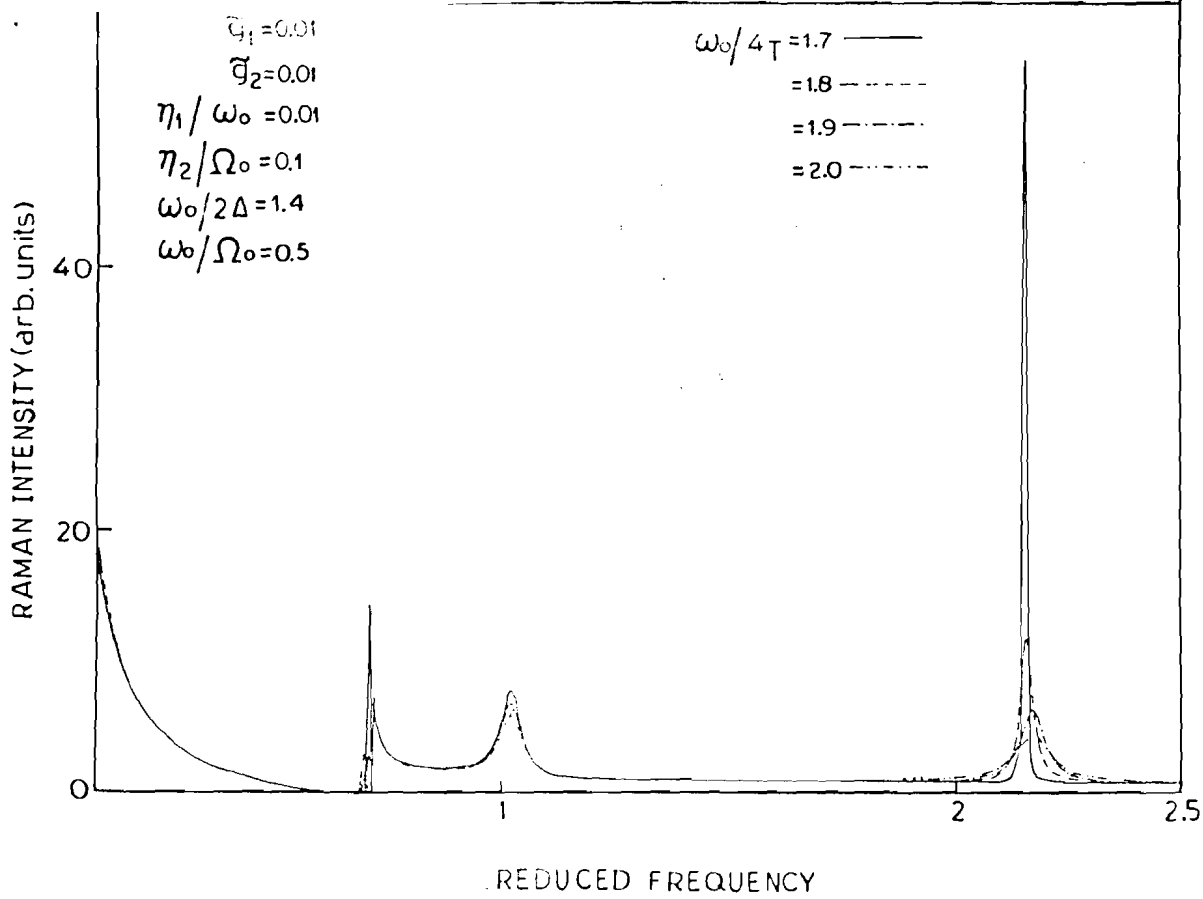
η_2/Ω_0=0.1
ω_0/Ω_0=2.0
$\omega_0/4_T$=1.2
=1.4
=1.6
=1.8
=2.0
RAMAN INTENSITY (arb. units)
REDUCED FREQUENCY
0
20
40
60
1
2
2.5

Fig.1(b)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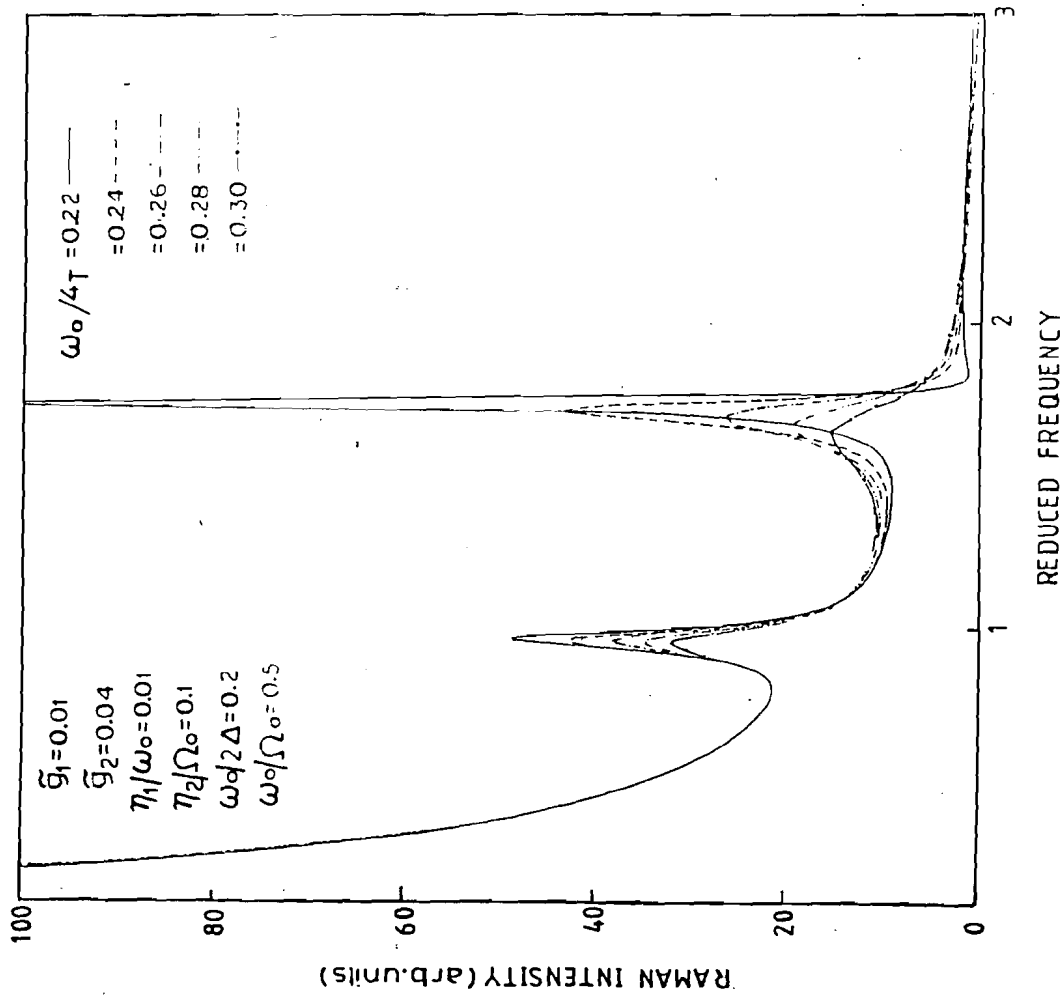
$\tilde{g}_2$=0.01
η_1/ω_0 =0.01
η_2/Ω_0 =0.1
$\omega_0/2\Delta$ =1.4
ω_0/Ω_0=0.5
$\omega_0/4_T$ =1.7
=1.8
=1.9
=2.0
RAMAN INTENSITY (arb. units)
0
20
40
1
2
2.5
REDUCED FREQUENCY

Fig.2(b)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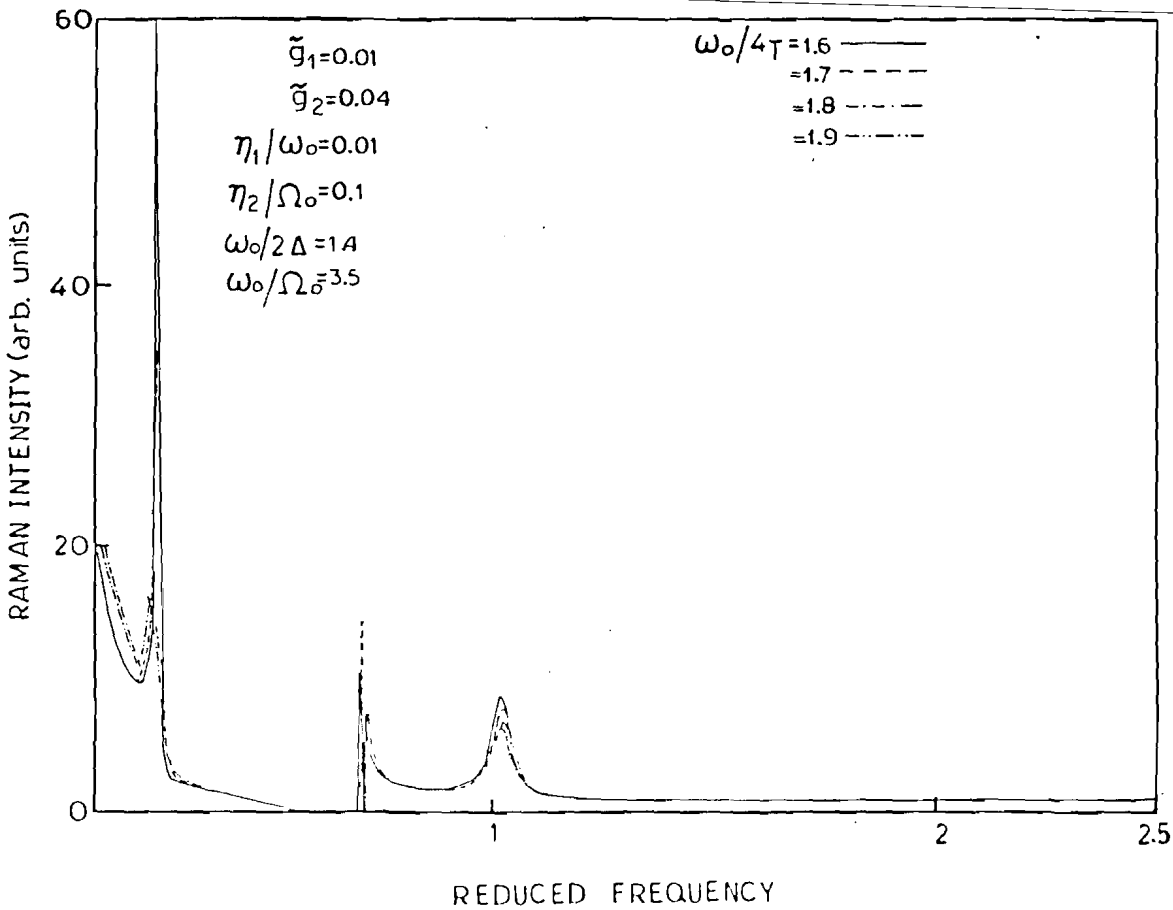
η_1/ω_0=0.01
η_2/Ω_0=0.1
$\omega_0/2\Delta$=0.2
ω_0/Ω_0=0.5
$\omega_0/4_T$ =0.22
=0.24
=0.26
=0.28
=0.30
RAMAN INTENSITY (arb.units)
0
20
40
60
80
100
1
2
3
REDUCED FREQUENCY

Fig.2(a)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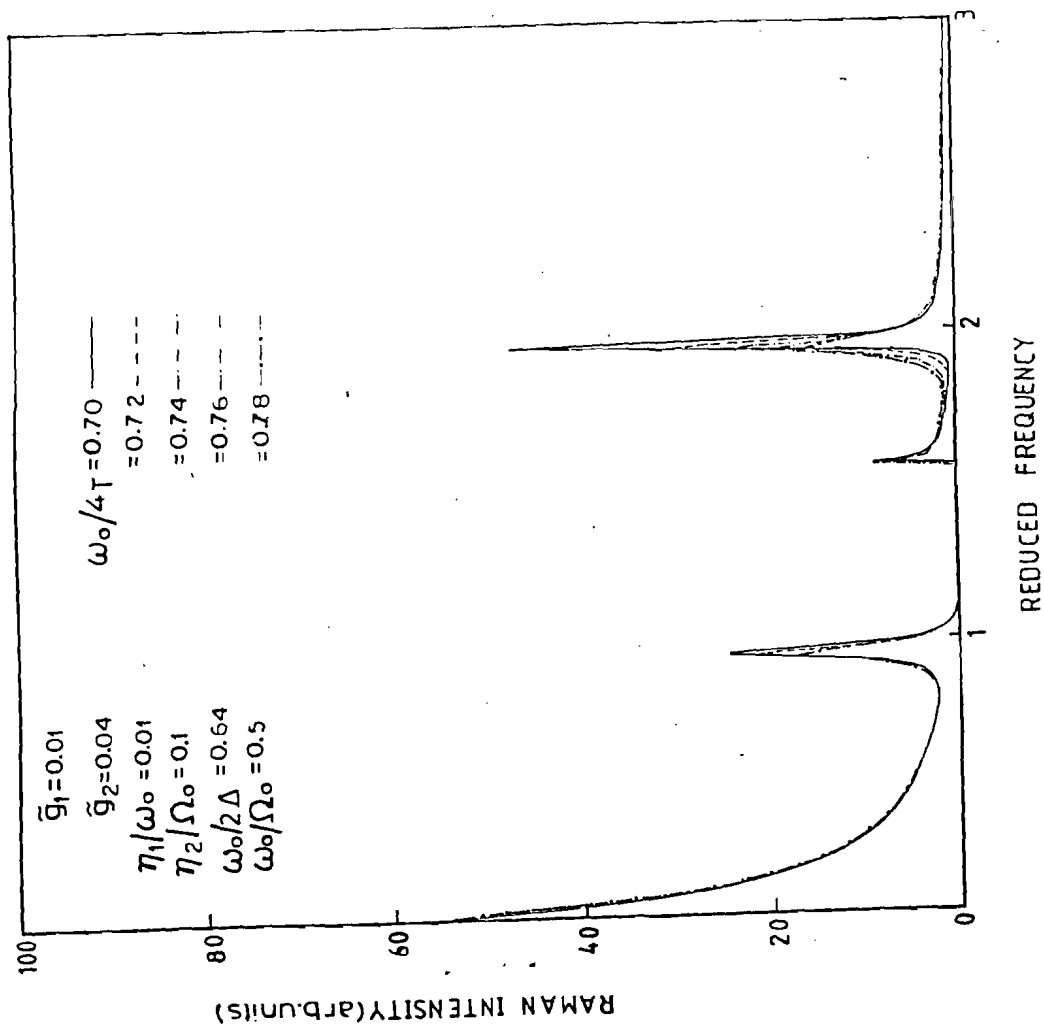
$\tilde{g}_2$=0.04
η_1/ω_o=0.01
η_2/Ω_o=0.1
$\omega_o/2\Delta$=1.4
ω_o/Ω_δ=3.5
$\omega_o/4_T$=1.6
=1.7
=1.8
=1.9
RAMAN INTENSITY (arb. units)
REDUCED FREQUENCY
0
20
40
60
1
2
2.5

Fig.2(d)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$\tilde{g}_2$=0.04
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$\omega_o/2\Delta$ =0.64
ω_o/Ω_o =0.5
$\omega_o/4_T$=0.70
=0.72
=0.74
=0.76
=0.78
RAMAN INTENSITY (arb.units)
REDUCED FREQUENCY
0
20
40
60
80
100
1
2
3

Fig.2(c)