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LO-PHONON RAMAN LINE IN MAGNETIC FIELD(*)

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ABSTRACT

Raman scattering by LO-phonons in the presence of a constant magnetic field is considered. The effect of the electron-phonon interaction on the Raman line is studied. It is shown that significant effects appear when, for certain experimental geometries, a harmonic of the cyclotron frequency equals the LO-phonon frequency. The frequency shift and line shape are discussed and application to GaAs is made.

ABSTRACT

Nous etudions l'effet d'interaction electron-fonon sur la ligne Raman dans la diffusion Raman de fonons optiques longitudinales en presence d'un champ magnetique constant. Nous montrons que des effets significatifs apparaissent quant, pour des determinées geometries experimentales, un harmonique de la frequence de ciclotron est egale a celle du fonon longitudinale. On a fait une etude du deplacement et de la forme de la ligne Raman et une aplication au cas du GaAs.

INELASTIC scattering of electro-magnetic radiation by crystals subjected to a constant magnetic field has recently been considered by several authors, who studied either scattering by the cyclotron levels[1] or by the coupled electron-phonon system.[2,3] Genkin and Zil'berberg [2] have shown that an enhanced Raman peak due to scattering by LO-phonons of frequency ω_o will be observed, when ω_o equals a harmonic of the cyclotron frequency ω_c. In the same conditions an additional enhancement of the LO-phonon Raman line can also be observed, which is assumed to be due to an LO-phonon distribution strongly departed from equilibrium.[4]

Raman scattering experiments can be a very convenient way to study the spectrum of the LO-phonon-cyclotron system in conditions of approximate statistical equilibrium or in the presence of "hot excitations". In order to get the proper information from the experimental data, a detailed theoretical study needs to be available. In this letter we present an extension of the works of references (2) and (3). We study the influence of the electron-LO-phonon interaction on the LO-phonon Raman line and find that significant effects appear when the resonant condition $\omega_o = n\omega_c$ (n integer) is satisfied.

We consider an n-type semiconductor in a constant magnetic field $\underset{\sim}{H}$ described by a vector potential $\underset{\sim}{A}$ given in the Landau gauge $\underset{\sim}{A} \equiv (-yH,0,0)$. Within the effective mass approximation the one-electron wave functions are the Landau functions $|nk_xk_z\sigma\rangle$.[5] The electron-phonon interaction will be given by the Fröhlich Hamiltonian

$$H_{EP} = \sum_{\tilde{q}} \sum_{nn'} \sum_{\sigma \tilde{k}\tilde{k}'} A_{\tilde{q}} I_{nn'}(\tilde{q}) C^{+}_{n\tilde{k}\sigma} C_{n'\tilde{k}'\sigma} (b_q + b^{+}_{-q}) \qquad (1)$$

where

$$\left| A_{\tilde{q}} \right|^2 = \frac{e^2}{q^2} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_o} \right) \frac{2\pi\hbar\omega_o}{V}$$

and

$$|I_{nn'}(\tilde{q})|^2 = \frac{n'!}{n!} (\lambda^2 q_\perp^2 / 2)^{n-n'} e^{-\lambda^2 q_\perp^2 / 2} \left| L^{n-n'}_{n'} (\lambda^2 q_\perp^2 / 2) \right|^2$$

where $n > n'$. In above expressions λ is the magnetic length $\lambda^2 = \hbar c/eH$, $q_\perp$ is the modulus of the component of $\tilde{q}$ normal to $\tilde{H}$, and L^m_n are Laguerre functions. The rest of the nomenclature is standard, except for $\tilde{k}$ which represents the vector of components k_x and k_z .

Using van Hove's method,[6] in conjunction with the Green's function formalism described in the already classical paper by Zubarev,[7] one finds, as in ref.(3), the differential Raman scattering cross section for LO-phonons

$$I(\omega) = \frac{d^2\sigma}{d\Omega d\omega} / \left(\frac{d\sigma}{d\Omega}\right)_o = Im\, G_{\tilde{q}}(\omega + is) \qquad (2)$$

where $(d\sigma/d\Omega)_o$ is the scattering amplitude as given by Genkin and Zil'berberg [2] and $G_{\tilde{q}}(\hbar^{-1}E)$ is the one-phonon Green's function [7] $\langle\langle b_q \mid b^+_q ; \hbar^{-1}\tilde{E} \rangle\rangle$. It should be born in mind that $(d\sigma/d\Omega)_o$ has resonant denominators and therefore it requires a more careful treatment, which should include lifetimes on the intermediate states.

Next we proceed to evaluate $G_{\tilde{q}}(\hbar^{-1}E)$ including Fröhlich interaction (1). Using the standard decoupling procedures [7] one finds

$$G_{\underset{\sim}{q}}(\hbar^{-1}E) = \frac{1}{2\pi} / [\hbar^{-1}E - \omega_o - P_{\underset{\sim}{q}}(\hbar^{-1}E)] \tag{3}$$

where

$$P_{\underset{\sim}{q}}(\hbar^{-1}E) = 2\hbar^{-1} \sum_{nn'} \sum_{\underset{\sim}{k}} |A_{\underset{\sim}{q}}|^2 |I_{nn'}(\underset{\sim}{q})|^2 \frac{f(n,\underset{\sim}{k}-\underset{\sim}{q}) - f(n',\underset{\sim}{k})}{E-\varepsilon(n',\underset{\sim}{k})+\varepsilon(n,\underset{\sim}{k}-\underset{\sim}{q})} \tag{4}$$

with $f(n,\underset{\sim}{k})$ the Fermi-Dirac distribution functions, and $\varepsilon(n,\underset{\sim}{k}) = (n + 1/2)\hbar\omega_c + \frac{\hbar^2 k_z^2}{2m} - \mu$. Here μ is the chemical potential of the electron gas. We consider of the case of concentrations n_e such that the dependence of μ on the magnetic field can be neglected and then $\mu = \frac{\hbar^2}{2m}(3\pi^2 n_e)^{2/3}$. When $\hbar^{-1}E=\omega+is$, with $s \longrightarrow +0$, as is necessary in Eq. (2), one finds for the real part, $P_{\underset{\sim}{q}}(\omega)$, and the imaginary part, $\gamma_{\underset{\sim}{q}}(\omega)$, of $P_{\underset{\sim}{q}}(\omega + is)$ at $T = 0^oK$

$$P_{\underset{\sim}{q}}(\omega) = \sum_{n=0}^{n_o} \sum_{\ell=1}^{\infty} \frac{C(q_\perp,q_z)}{\pi} \frac{\rho^{\ell-1}}{(\ell!)^2} \frac{(n+\ell)!}{n!} \times$$

$$\left\{ \ell n \left| \frac{\omega^2 - (\ell\omega_c - \beta^{(n+\ell)})^2}{\omega^2 - (\ell\omega_c + \beta^{(n+\ell)})^2} \right| + \right.$$

$$\left. \ell n \left| \frac{\omega^2 - (\ell\omega_c + \beta^{(n)})^2}{\omega^2 - (\ell\omega_c - \beta^{(n)})^2} \right| \right\}$$

and

$$\gamma_{\underset{\sim}{q}}(\omega) = \sum_{n=0}^{n_o} \sum_{\ell=1}^{\infty} \gamma_{n\ell}(\omega) \tag{6}$$

where

$$\gamma_{n\ell}(\omega) = C(q_\perp, q_z) \frac{n!}{(n+\ell)!} \left| \binom{n+\ell}{n} \right|^2 \rho^{\ell-1} \tag{6a}$$

if $\beta^{(n+\ell)} < | \omega - \ell\omega_c | < \beta^{(n)}$ and zero otherwise.

In Eqs. (5) and (6) we have introduced

$$\rho = \lambda^2 q_\perp^2 / 2 \quad , \quad C(q_\perp, q_z) = \frac{e^2 m\omega_o q_\perp^2}{\hbar^2 q_z (q_\perp^2 + q_z^2)} \left(\frac{1}{\varepsilon_\infty} - \frac{1}{\varepsilon_o}\right)$$

and $\beta^{(n)} = \hbar q_z k_{Fz}^{(n)} / m$, where $k_{Fz}^{(n)\,2} = \frac{2m}{\hbar^2} [\mu - (n+1/2)\hbar\omega_c]$ for $n \leq n_o$ and zero for $n > n_o$, where n_o is the number of the highest non-empty Landau sub-band. Since we are interested in $\underset{\sim}{q}$ (the photon momentum transfer) very small we have assumed that the experimental conditions are such that $\rho << 1$ and $q_z << k_{Fz}^{(n)}$. Blank and Kaner [8] derived expressions for $P_{\underset{\sim}{q}}(\omega)$ and $\gamma_{\underset{\sim}{q}}(\omega)$ for acoustic phonons interacting with cyclotron modes. In their calculation the leading contributions are due to electronic intra-level transitions whereas in our calculation, involving optical phonons, electronic inter-level transitions are the most important contributions.

We proceed to specialize our results to the case of GaAs. The following values are used $m = 0.068m_o$, $E_G = 1.52$ eV , $\alpha = 0.06$, $\varepsilon_\infty = 10.9$, $\varepsilon_o = 12.9$, $\omega_o = 297\ cm^{-1}$, and $n_e = 5.7 \times 10^{17}\ cm^{-3}$.

We consider the $\lambda_L = 10150\ \mathring{A}$ YAG line, which produces maximum q values of roughly $2\times10^5 cm^{-1}$. With these values the resonant condition $n\omega_c \sim \omega_o$ occurs at values of $H = \frac{217}{n}$ kOe.

It is found that for $n = 1$ the damping function $\gamma_{\underset{\sim}{q}}(\omega_c)$ is very large, nearly ten times ω_o, which raises doubts about the applicability of our results in this case. Furthermore, hybridization of the $n = 1$ mode with plasma modes should also be included in the calculation. For these reasons, we concentrate our attention on the $n = 2$ peak when $\gamma_{\underset{\sim}{q}}(2\omega_c)$, for not too small values of the angle θ between $\underset{\sim}{q}$ and the normal to the magnetic field, is a fraction of ω_o. A constant value Γ_o is added to $\gamma_{\underset{\sim}{q}}(\omega)$ in order to incorporate relaxation effects due to imperfections and impurities in the sample.

In Figure 1 we have the relaxation function $\gamma(\omega)$ for $\omega_c/\omega_o = 0.5$. We notice the "ladder-type" form of this function, which should be evident in the shape of the spectra. In Figure 2, we present the Raman spectra derived from Eqs.(2)-(6), for $\theta=20^o$ with (a) $\omega_c/\omega_o = 0.43$ and (b) $\omega_c/\omega_o = 0.5$. The double structure of the spectra is obtained, as should be expected.[3] In case (a) one finds a small peak near $2\omega_c$ and the strongest near ω_o. In case (b) both peaks are overlapped with center near ω_o. Similarly to the case of scattering by the electronic excitations,[3] the "phonon strength" takes its larger value on the upper mode for $2\omega_c < \omega_o$ and in the lower mode for $2\omega_c > \omega_o$.

The frequency dependence of the relaxation function $\gamma_{\underset{\sim}{q}}(\omega)$ produces a distortion in the otherwise Lorentzian line and introduces a complicated structure which, however, could be softened in actual experimental circumstances by temperature effects and other damping effects not taken into account in our model.

Finally, it should be observed that the present calculation shows that the peak values of $I(\omega)$ are an increasing function of n , as opposed to $(d\sigma/d\Omega)_o$ which is a decreasing function of n.(2)

In conclusion, we may say that electron-phonon interaction largely affects the line-shapes and peak values of Raman lines in magneto-Raman scattering experiments in doped semiconductors.These effects together with the resonant effects (2) in the scattering amplitude $(d\sigma/d\Omega)_o$ and non-equilibrium conditions (4) should be carefully considered when analyzing experimental data.

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FIGURE CAPTION

Figure 1: Real, $P(\omega)$, and Imaginary, $\gamma(\omega)$, parts of the polarization operator $P_{\tilde{q}}(\omega)$ of Eq. (4) for GaAs. ($\omega_c/\omega_o = 0.5$)

Figure 2: The LO-phonon Raman line at $T = 0^oK$ for two different ω_c/ω_o ratios.

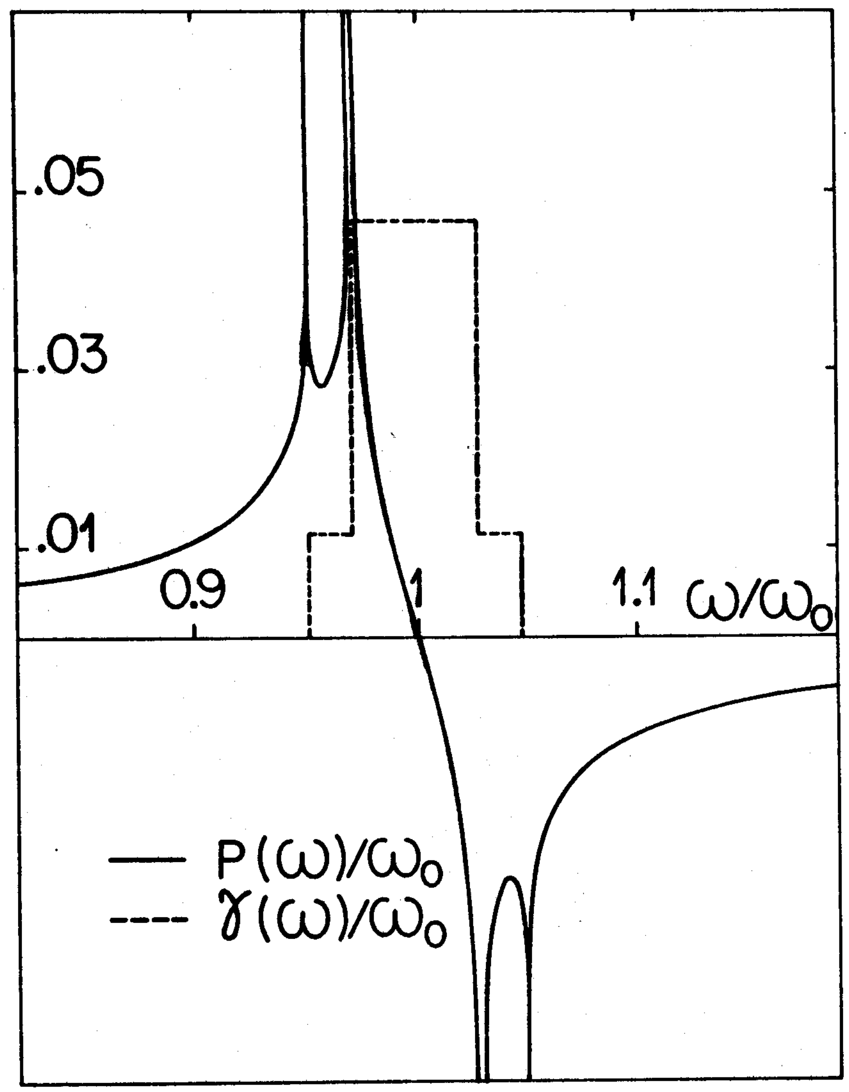

Fig. 1

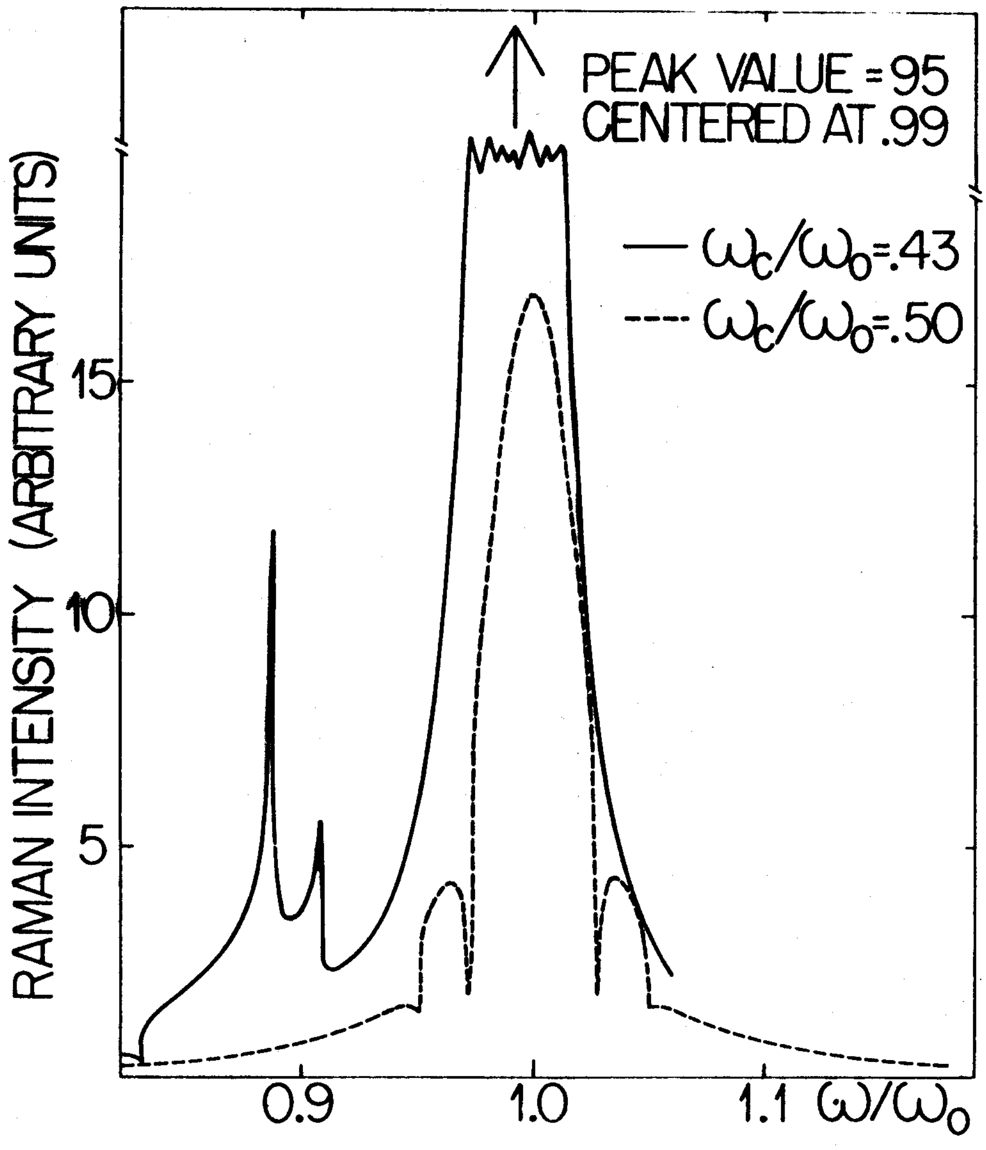
PEAK VALUE = 95
CENTERED AT .99
ω_c/ω_0=.43
ω_c/ω_0=.50
RAMAN INTENSITY (ARBITRARY UNITS)
15
10
5
0.9
1.0
1.1 ω/ω_0

Fig. 2