Optically detected magnetophonon resonances in GaAs

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Magnetophonon resonances are found for $\omega_c = \omega_{\mathrm{LO}}/N$ with $N = 1,2,3\ldots$ in the polaron cyclotron resonance (CR) linewidth and effective mass of bulk polar semiconductors. The CR mass and the linewidth are obtained from the full polaron magneto-optical absorption spectrum which are calculated using the memory function technique. The amplitude of the resonant peak in the linewidth can be described by exponential law at low temperature. [S0163-1829(99)09139-0]

I. INTRODUCTION

Magnetophonon resonance (MPR) occurs when two Landau levels are a phonon energy apart that leads to a resonant scattering due to emission or absorption of phonons. Since the pioneer work by Gurevich and Firsov,[1] this effect has been extensively studied in bulk[2,3] as well as low-dimensional semiconductor systems.[4–9] The resonant character makes it a powerful spectroscopic tool. Magnetophonon resonances have been used to obtain information on band-structure parameters, such as the effective mass and the energy levels, and on the electron-phonon interaction. The vast majority of work on the MPR has been done on the transport properties of semiconductors, usually the magnetoresistance, which inevitably involves a complicated average of scattering processes. The oscillations in the magnetoresistance are the results of a combination of scattering and broadening processes that can lead to a quite complicated dependence of the resonance amplitudes on doping, sample structure, carrier concentration, and temperature. However, the MPR can also be observed directly through a study of the electron cyclotron resonance (CR) linewidth and effective mass, i.e., the so-called optically detected MPR (ODMPR), as was demonstrated in two-dimensional (2D) semiconductor systems of $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ heterojunctions by Barnes *et al.*[10] The ODMPR allows one to make quantitative measurements of the scattering strength for specific Landau levels and yields direct information on the nature of the electron-phonon interaction in semiconductors.

In this work, we extend the theory for ODMPR to three-dimensional (3D) systems and present a theoretical study of the magnetophonon resonances in the frequency-dependent conductivity in bulk polar semiconductors. Our calculations show strong oscillations of both the linewidth and the effective mass in a 3D system of GaAs that indicate that the ODMPR should also be observed experimentally in bulk polar semiconductors.

The present paper is organized as follows. In Sec. II, we present our theoretical formulations of the problem. The numerical results and discussions are given in Sec. III, and we summarize our results in Sec. IV.

II. THEORETICAL FRAMEWORK

Magnetophonon resonance is essentially a single-particle effect and, consequently, can be treated as a one-polaron problem. We consider a polar semiconductor in a uniform magnetic field **B** directed along the z axis. The system under consideration can be described by the following Hamiltonian,

$$H = H_e + H_{\mathrm{ph}} + H_{\mathrm{int}} \tag{1}$$

with

$$H_e = \frac{1}{2m_b}(\vec{p} + e\vec{A})^2 \tag{2}$$

and

$$H_{\mathrm{ph}} = \sum_{\vec{q}} \hbar\omega_{\vec{q}}(a^\dagger_{\vec{q}} a_{\vec{q}} + \tfrac{1}{2}), \tag{3}$$

where m_b is the bare electron effective mass, the vector potential $\vec{A} = B/2(-y,x,0)$ is chosen in the symmetrical Coulomb gauge, $\vec{q}$ $(\vec{r})$ the momentum (position) operator of the electron, $a^\dagger_{\vec{q}}$ $(a_{\vec{q}})$ the creation (annihilation) operator of an optical phonon with wave vector $\vec{q}$ and energy $\hbar\omega_{\vec{q}}$. The electron-phonon interaction Hamiltonian H_{int} is given by the Fröhlich interaction Hamiltonian

$$H_{\mathrm{int}} = \sum_{\vec{q}} (V_{\vec{q}} a_{\vec{q}} e^{i\vec{q}\cdot\vec{r}} + V^*_{\vec{q}} a^\dagger_{\vec{q}} e^{-i\vec{q}\cdot\vec{r}}), \tag{4}$$

where

$$V_{\vec{q}} = -i\hbar\omega_{\mathrm{LO}}\left(\frac{\hbar}{2m_b\omega_{\mathrm{LO}}}\right)^{1/4}\sqrt{\frac{4\pi\alpha}{Vq^2}}, \tag{5}$$

and α is the electron-LO-phonon coupling constant.

First, we calculate the optical-absorption spectrum of the polaron in magnetic fields from which we are able to investigate the polaron CR spectrum and the MPR effects. For convenience we use units such that $\hbar = m_b = \omega_{\mathrm{LO}} = 1$. Within the linear-response theory, the frequency-dependent magneto-optical-absorption spectrum for cyclotron resonance[11–13] is given by

$$A(\omega)=-\frac{1}{2}\frac{\mathrm{Im}\,\Sigma(z)}{[\omega-\omega_c-\mathrm{Re}\,\Sigma(z)]^2+[\mathrm{Im}\,\Sigma(z)]^2}, \tag{6}$$

where $\omega_c=eB/m_b$ is the unperturbed electron cyclotron frequency, $\Sigma(z)$ is the so-called memory function, and $z=\omega+i\gamma$ and γ is a broadening parameter. Notice that γ is introduced semiempirically to remove the divergence of the Landau-level density of states. We take γ as a constant. For the magneto-optical-absorption spectrum in the Faraday (active-mode) configuration, which corresponds to the cyclotron resonance experiments, the memory function is given by[12]

$$\Sigma(z)=\frac{1}{2m_b}\sum_{\vec{q}} q^2|V_{\vec{q}}|^2F_{\vec{q}}(z) \tag{7}$$

with

$$F_{\vec{q}}(z)=-\frac{2}{z}\int_0^\infty dt(1-e^{izt})\mathrm{Im}\langle[b_{\vec{q}}(t),b_{\vec{q}}^{\dagger}(0)]\rangle, \tag{8}$$

where $b_{\vec{q}}=a_{\vec{q}}e^{i\vec{q}\cdot\vec{r}}$, and the correlation function is given by

$$\langle[b_{\vec{q}}(t),b_{\vec{q}}^{\dagger}(0)]\rangle=[1+n(\omega_{\mathrm{LO}})]e^{-i\omega_{\mathrm{LO}}t}S^*(-\vec{q},t)-n(\omega_{\mathrm{LO}})e^{-i\omega_{\mathrm{LO}}t}S(\vec{q},t), \tag{9}$$

where

$$n(\omega_{\mathrm{LO}})=\frac{1}{e^{\beta\omega_{\mathrm{LO}}}-1} \tag{10}$$

is the number of the LO phonons and

$$S(\vec{q},t)=\langle e^{i\vec{q}\cdot\vec{r}(t)}e^{-i\vec{q}\cdot\vec{r}(0)}\rangle \tag{11}$$

is the space Fourier transform of the electron density-density correlation function. In Eq. (10) $\beta=1/k_BT$, where k_B is the Boltzmann constant. For a weak electron-LO-phonon coupling system, i.e., $\alpha\ll 1$, the density-density correlation function is calculated for a free electron in a magnetic field which is given by

$$S(\vec{q},t)=e^{q_z^2D(t)}e^{-q^2D_H(t)} \tag{12}$$

with

$$D(t)=\frac{1}{2}(-it+t^2/\beta) \tag{13}$$

and

$$D_H(t)=\frac{1}{2\omega_c}[1-e^{i\omega_ct}+4n(\omega_c)\sin^2(\omega_ct/2)].$$

From the above equations, we obtain the memory function for $\gamma=0$. The calculation proceeds along the lines of a similar calculation which was presented in Ref. 12. The results for the memory function are

$$\begin{aligned}\mathrm{Re}\,\Sigma(\omega)=&\frac{\alpha\sqrt{\beta}}{2\pi\omega}\frac{\omega_c\tanh(\beta\omega_c/2)}{\sinh(\beta/2)}\sum_{n,n'=0}^{\infty}\frac{[2\cosh(\beta\omega_c/2)]^{-(n+n')}}{n!n'!}\int_0^\infty\frac{dx}{x}E_{n+n'+1}\left(\frac{x^2}{\omega_c\tanh(\beta\omega_c/2)}\right)\\&\times\left\{\exp\left(\frac{\beta\omega_{nn'}}{2}\right)\left[2D\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}\omega_{nn'}\right)-D\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{nn'}+\omega)\right)-D\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{n'n}-\omega)\right)\right]\right.\\&+\exp\left(-\frac{\beta\omega_{n'n}}{2}\right)\left[2D\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}\omega_{n'n}\right)\right.\\&\left.\left.-D\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}-\omega)\right)-D\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}+\omega)\right)\right]\right\},\end{aligned} \tag{14}$$

and

$$\begin{aligned}\mathrm{Im}\,\Sigma(\omega)=&-\frac{\alpha\sqrt{\beta}}{4\sqrt{\pi}\omega}\frac{\omega_c\sinh(\beta\omega/2)\tanh(\beta\omega_c/2)}{\sinh(\beta/2)}\sum_{n,n'=0}^{\infty}\frac{[2\cosh(\beta\omega_c/2)]^{-(n+n')}}{n!n'!}\\&\times\int_0^\infty\frac{dx}{x}E_{n+n'+1}\left(\frac{x^2}{\omega_c\tanh(\beta\omega_c/2)}\right)\\&\times\left[\exp\left(-\frac{\beta x}{4}-\frac{\beta}{4x}(\omega_{nn'}-\omega)\right)+\exp\left(-\frac{\beta x}{4}+\frac{\beta}{4x}(\omega_{n'n}-\omega)\right)\right],\end{aligned} \tag{15}$$

where $\omega_{nn'}=1+(n-n')\omega_c$, $D(x)$ is the Dawson's integral function, and

$$E_n=\int_0^\infty dt\frac{t^ne^{-t}}{t+x}. \tag{16}$$

In the case of $\gamma\neq 0$, the calculation is more tedious. We obtain the following results of the memory function,

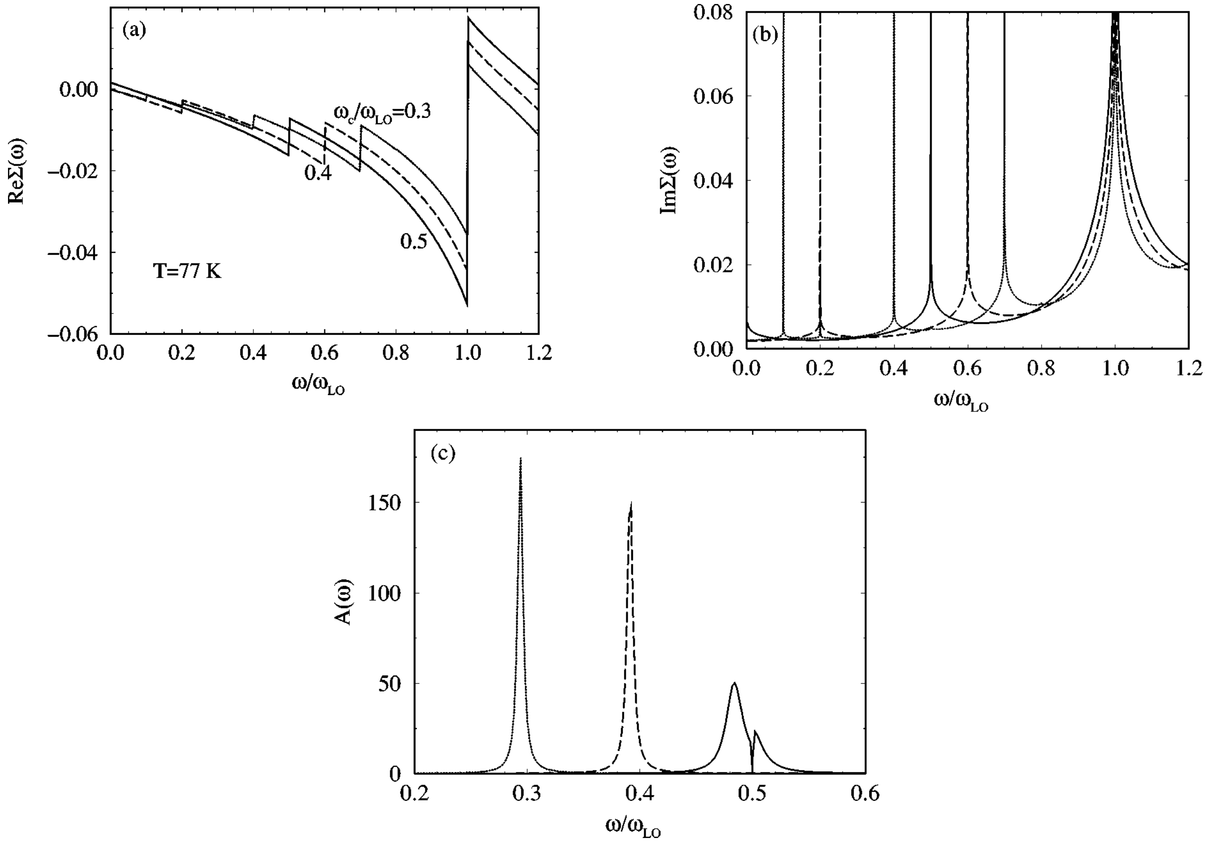

FIG. 1. (a) $\mathrm{Re}\,\Sigma(\omega)$ and (b) $\mathrm{Im}\,\Sigma(\omega)$ as a function of frequency ω in GaAs at different magnetic fields $\omega_c/\omega_{\mathrm{LO}}=0.3$ (dotted curves), 0.4 (dashed curves), and 0.5 (solid curves). The corresponding absorption spectra are given in (c). The broadening parameter $\gamma=0$ and temperature $T=77$ K.

$$\mathrm{Re}\,\Sigma(\omega)=-\frac{\alpha}{\sqrt{2}\,\pi(\omega^2+\gamma^2)}[\omega I_1(\omega)+\gamma I_2(\omega)] \tag{17}$$

and

$$\mathrm{Im}\,\Sigma(\omega)=\frac{\alpha}{\sqrt{2}\,\pi(\omega^2+\gamma^2)}[\omega I_2(\omega)+\gamma I_1(\omega)] \tag{18}$$

with

$$\begin{aligned}
I_1(\omega)=&-\sqrt{2\beta}\,\frac{\omega_c\tanh(\beta\omega_c/2)}{\sinh(\beta/2)}\sum_{n,n'=0}^{\infty}\frac{[2\cosh(\beta\omega_c/2)]^{-(n+n')}}{n!n'!}\int_0^{\infty}\frac{dx}{x}E_{n+n'+1}\left(\frac{x^2}{\omega_c\tanh(\beta\omega_c/2)}\right)\\
&\times\left(\exp\left(\frac{\beta\omega_{nn'}}{2}\right)\left\{D\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}\omega_{nn'}\right)\right.\right.\\
&-\frac{\sqrt{\pi}}{4}\left[\mathrm{Im}\,W\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{nn'}+\omega+i\gamma)\right)+\mathrm{Im}\,W\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{n'n}-\omega+i\gamma)\right)\right]\Bigg\}\\
&+\exp\left(-\frac{\beta\omega_{n'n}}{2}\right)\left\{D\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}\omega_{n'n}\right)\right.\\
&\left.\left.-\frac{\sqrt{\pi}}{4}\left[\mathrm{Im}\,W\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}-\omega-i\gamma)\right)+\mathrm{Im}\,W\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}+\omega-i\gamma)\right)\right]\right\}\right),
\end{aligned} \tag{19}$$

and

$$I_2(\omega) = -\frac{\sqrt{\pi\beta}}{2\sqrt{2}} \frac{\omega_c \tanh(\beta\omega_c/2)}{\sinh(\beta/2)} \sum_{n,n'=0}^{\infty} \frac{[2\cosh(\beta\omega_c/2)]^{-(n+n')}}{n!n'!} \int_0^\infty \frac{dx}{x} E_{n+n'+1}\left(\frac{x^2}{\omega_c \tanh(\beta\omega_c/2)}\right)$$
$$\times\left\{\exp\left(\frac{\beta\omega_{nn'}}{2}\right)\left[\mathrm{Re}\,W\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{nn'}+\omega+i\gamma)\right)-\mathrm{Re}\,W\left(\frac{\sqrt{\beta}x}{2}+\frac{\sqrt{\beta}}{2x}(\omega_{nn'}-\omega+i\gamma)\right)\right]\right.$$
$$\left.+\exp\left(-\frac{\beta\omega_{n'n}}{2}\right)\left[\mathrm{Re}\,W\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}-\omega-i\gamma)\right)-\mathrm{Re}\,W\left(\frac{\sqrt{\beta}x}{2}-\frac{\sqrt{\beta}}{2x}(\omega_{n'n}+\omega-i\gamma)\right)\right]\right\}, \quad (20)$$

where $W(z)=e^{-z^2}\mathrm{erfc}(-iz)$ is the complex error function.

III. NUMERICAL RESULTS AND DISCUSSIONS

In this section, we are going to present our numerical results on the magneto-optical-absorption spectra and to study the magnetophonon resonant effects. As an example of weak electron-LO-phonon coupling, we apply our theory to semiconductor GaAs where $\alpha=0.07$. First, we show some numerical results for temperature $T=77$ K and level broadening parameter $\gamma=0$. Due to the importance of the memory function in the absorption spectrum, we plot the real and imaginary parts of the memory function in Figs. 1(a) and 1(b), respectively, as a function of frequency at different magnetic fields. We see that, at $\omega=|\omega_{\mathrm{LO}}-n\omega_c|$ ($n=0,1,2,\ldots$), $\mathrm{Re}\,\Sigma(\omega)$ exhibits a jump while $\mathrm{Im}\Sigma(\omega)$ diverges logarithmically. The discontinuity of $\mathrm{Re}\Sigma(\omega)$ and the divergency in $\mathrm{Im}\Sigma(\omega)$ reflects the resonant coupling between the state $E_0+\omega_{\mathrm{LO}}$ and Landau level $E_n=(1/2+n)\omega_c$. The stronger this coupling, the larger the discontinuity in $\mathrm{Re}\Sigma(\omega)$. Actually, the real part of the memory function $\mathrm{Re}\Sigma(\omega)$ is responsible for the shift in the observed CR energy which is due to the electron-phonon interaction, while the imaginary part leads to a broadening of the spectrum which is a result of scattering. When $\mathrm{Im}\Sigma(\omega)=0$ like in a 2D system, the absorption is a δ function, and its position is determined by the equation $\omega_c^*-\omega_c-\mathrm{Re}\,\Sigma(\omega_c^*)=0$. Figure 1(b) shows that in the present system the $\mathrm{Im}\,\Sigma(\omega)$ is always non-zero, which reflects the 3D character of the electron states. The scattering in the direction parallel to the magnetic field results in a finite $\mathrm{Im}\,\Sigma(\omega)$ and, consequently, a finite linewidth even for $\gamma=0$. In Fig. 1(c), we show the corresponding magneto-optical-absorption spectra. The position of the absorption peak corresponds to the cyclotron resonant frequency ω_c^* at which the cyclotron resonance occurs. We see an asymmetric double peak structure around $\omega=\omega_{\mathrm{LO}}/2$ for $\omega_c=\omega_{\mathrm{LO}}/2$ (the solid curve), and the aborption becomes zero at $\omega=\omega_c=\omega_{\mathrm{LO}}/2$. The zeros in the absorption spectrum are a consequence of the divergences in $\mathrm{Im}\Sigma(\omega)$ and can be traced back to the divergent nature of the density of states. The double peak structure is a consequence of the magnetophonon resonance which leads to an anticross behavior in the CR spectrum. When the unperturbed CR frequency ω_c deviates from ω_{LO}/N, this splitting becomes very weak and difficult to be observed in the absorption spectrum. As we will see below, however, the magnetophonon resonance will strongly affect the linewidth of the magneto-optical absorption and the CR mass. From the dashed and dotted curves,

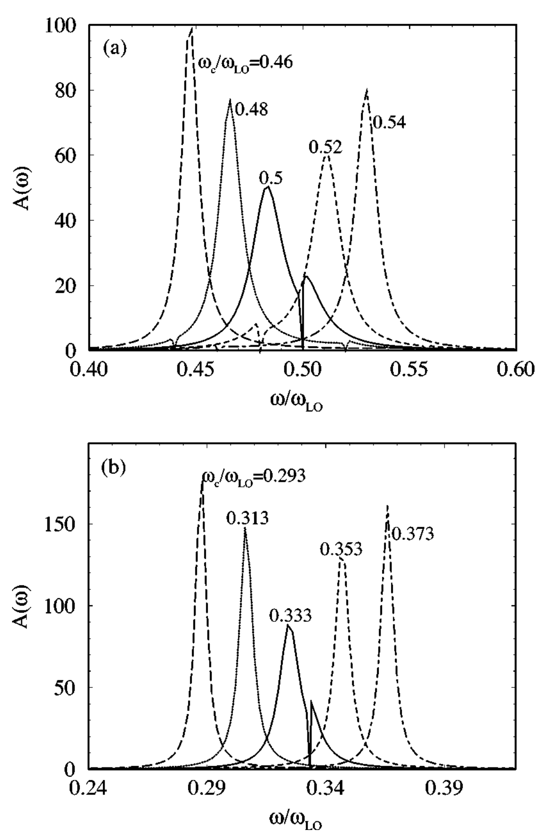

FIG. 2. The magneto-optical-absorption spectrum at around (a) $\omega_c/\omega_{\mathrm{LO}}=1/2$ and (b) $\omega_c/\omega_{\mathrm{LO}}=1/3$. $T=77$ K and $\gamma=0$.

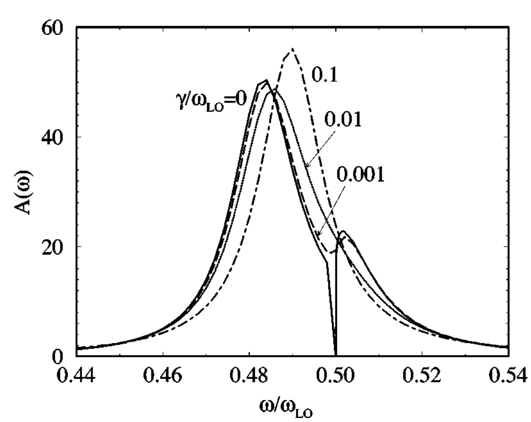

FIG. 3. The magneto-optical-absorption spectra as a function of frequency ω in GaAs for $\gamma/\omega_{\mathrm{LO}}=0$ (solid curve), 0.001 (dashed curve), 0.01 (dotted curve), and 0.1 (dash-dotted curve) at $\omega_c/\omega_{\mathrm{LO}}=0.5$ and $T=77$ K.

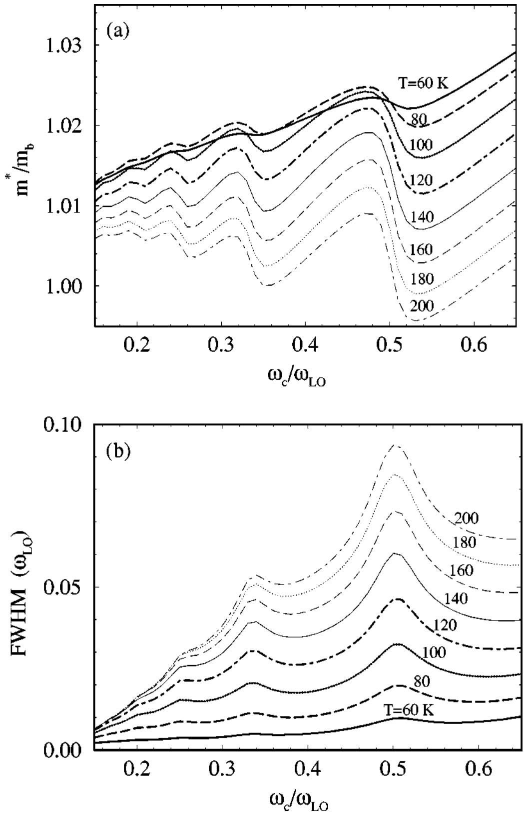

FIG. 4. (a) Polaron CR mass and (b) FWHM as a function of ω_c at different temperatures from 60 K to 200 K with $\gamma/\omega_{LO}=0.05$.

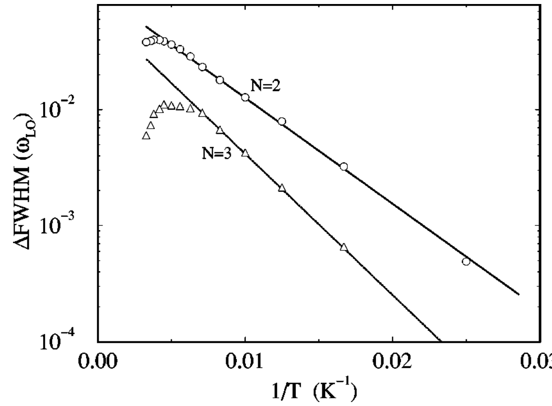

FIG. 6. An activation plot of the amplitude of the resonant peak in the FWHM at $\omega_c/\omega_{LO}=1/2$ (circles) and $\omega_c/\omega_{LO}=1/3$ (triangles) as a function of T^{-1}. The solid line $\propto \exp(-\hbar\omega_{LO}/2k_BT)$ and the dotted line $\propto \exp(-2\hbar\omega_{LO}/3k_BT)$.

we observe that the absorption peak appears at $\omega_c^* < \omega_c$ due to the polaron effect which shifts the cyclotron frequency to lower frequencies. The latter is often interpreted as an increase of the cyclotron mass, i.e., $\omega_c^* = eB/m^*$. In Fig. 2, we show the absorption spectrum around (a) $\omega_c=\omega_{LO}/2$ and (b) $\omega_c=\omega_{LO}/3$. The double peak structure disappears when ω_c deviates from ω_{LO}/N $(N=2,3)$. The absorption spectra also demonstrate clearly a nonlinear magnetic-field dependence of the peak position and linewidth around ω_{LO}/N.

Figure 3 demonstrates the effect of the broadening parameter γ on the absorption spectrum. Notice that, with increasing γ: (i) the double peak structure disappears for $\gamma > 0.01\omega_{LO}$, (ii) the zero in the absorption spectrum disappears when $\gamma > 0$, and (iii) the position of the absorption peak shifts to higher frequency. This indicates that the anticrossing behavior in the CR spectrum will be difficult to be observed experimentally at $\omega_c=\omega_{LO}/2$, due to broadening effects which are a consequence of scattering on e.g., impurities and acoustical phonons.

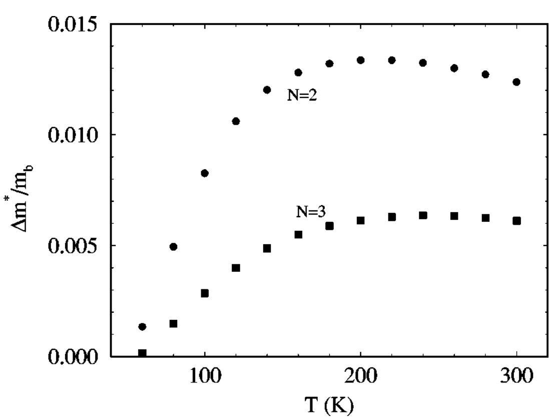

FIG. 5. The CR mass oscillation amplitude as a function of temperature at $\omega_c/\omega_{LO}=1/2$ (dots) and $\omega_c/\omega_{LO}=1/3$ (solid squares) with $\gamma/\omega_{LO}=0.05$.

As soon as the polaron CR frequency ω_c^* is determined from the position of the magneto-optical-absorption peak, the CR mass of the polaron is obtained by

$$m^*/m_b = \omega_c/\omega_c^* . \tag{21}$$

The numerical results of the polaron CR mass and the FWHM (full width at half maximum) for $\gamma=0.05\omega_{LO}$ are plotted as a function of the unperturbed CR frequency at different temperatures in Figs. 4(a) and 4(b), respectively. One observes that the polaron CR mass is an oscillatory function of magnetic field. Figure 4(b) shows that the FWHM of the polaron magneto-optical-absorption spectrum reach a local maximum at $\omega_c=\omega_{LO}/N$ where the polaron mass has an inflection point. This result demonstrates the derivativelike relation between the polaron CR mass and the linewidth which are due to the fact that the real and imaginary part of the memory function are related to each other through a Kramers-Kronig relation. One finds that, for temperature $T<100$ K, the resonance grows rapidly with increasing T. This effect can lead to a direct measure of the optical-phonon scattering rate. We also show an overall increase of the linewidth with temperature but an overall decrease of the effective mass for $T>80$ K. The resonant position is slightly larger than the unperturbed resonant condition $\omega_c=\omega_{LO}/N$ and is almost independent of temperature. A detailed analysis indicates that, at $N=2$ and 3, the peak position both in the FWHM and in the derivative of the CR mass is at about $0.504\omega_{LO}$ and $0.336\omega_{LO}$, respectively. Experimentally, this position determines the so-called fundamental field $B_0=m^*\omega_{LO}/e$, which is an important quantity to study the effective mass, nonparabolicity of the energy band, as well as the LO-phonon frequency. The linewidth is a direct measure of the lifetime of the state. Notice that the conventional MPR occurs in the resistivity, which is given by $\rho_{zz}=-\mathrm{Im}\Sigma(\omega=0)$. But ODMPR is related to both the real and imaginary part of the memory function which occurs for $\omega \neq 0$ and is a dynamical MPR.

Figure 5 shows the CR mass oscillation amplitude at $\omega_c/\omega_{\mathrm{LO}}=1/2$ and 1/3 as a function of temperature. With increasing temperature, the number of phonons increases and, consequently, the oscillation amplitude increases. On the other hand, the background electron-phonon scattering (coupling) increases, which results in a suppression of the oscillation amplitude. Figure 6 shows an activation plot of the amplitude of the resonant peak in the FWHM at $\omega_c/\omega_{\mathrm{LO}}=1/2$ and 1/3 as a function of T^{-1}. We find that, for the resonance around $N=2$, the linewidth can be described rather well by the exponential law $\exp(-\hbar\omega_{\mathrm{LO}}/2kT)$ for $T<240$ K, while that around $N=3$ can be described by $\exp(-2\hbar\omega_{\mathrm{LO}}/3kT)$ for $T<140$ K. This exponential behavior can be understood as follows. MPR is proportional to the number of LO phonons which are present and therefore should increase as $n(\omega_{\mathrm{LO}})$. On the other hand, thermal broadening of the Landau levels, which is proportional to $n(\omega_c)$, will diminish the resonant structure in ΔFWHM. Thus, this contribution decreases the resonant character, and consequently we expect that ΔFWHM $\sim n(\omega_{\mathrm{LO}})/n(\omega_c)\approx\exp[-\hbar(\omega_{\mathrm{LO}}-\omega_c)/k_BT]$ which agrees with the exponential laws found for $N=2$ and $N=3$.

IV. SUMMARY

We have extended the theory for ODMPR to three-dimensional (3D) systems and present the first detailed theoretical study of the magnetophonon resonance in the magneto-optical-absorption spectrum in bulk GaAs. In comparison to the corresponding 2D systems, the theoretically obtained amplitudes for the oscillations of both the linewidth and the effective mass in a 3D system are for GaAs predicted to be about half of those in 2D. Therefore, we believe that ODMPR can also be observed experimentally in bulk polar semiconductors. Our numerical results indicate that the amplitude of the resonant peak in the FWHM can be described by exponential law at low temperature.

ACKNOWLEDGMENTS

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