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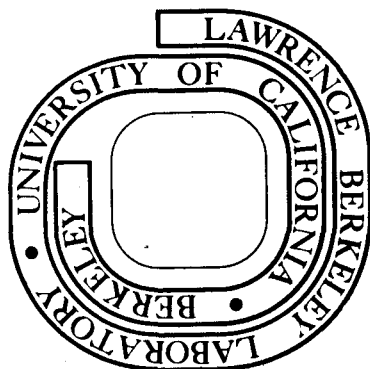
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TEMPERATURE DEPENDENCE OF THE BAND GAP AND COMPARISON WITH
THE THRESHOLD FREQUENCY OF PURE GaAs LASERS

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ABSTRACT

Recent measurements of the lasing energy as a function of temperature in high purity GaAs lasers, have attempted to investigate the relation between this energy and the one electron band gap. Because of a lack of precision in the position of the band gap at high temperature, these measurements show strongly conflicting results.

In this work, we report two sets of differential reflectivity measurements (electroreflectivity and piezoreflectivity) performed on GaAs under high resolution conditions. Both series of results give for the excitonic absorption edge at room temperature a value:

$E_0 = 1.424 \text{ eV} \pm 0.002 \text{ eV}$ which is about 20 meV higher than the lasing energy reported for the highest purity GaAs samples.

This result confirm that the lasing energy in GaAs is well below the one electron band gap. In addition, we show that this energy separation is an increasing function of temperature. Lastly we discuss

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a simple model of band to band recombination including electron-electron interaction effects and we show that it permits to calculate a temperature dependence of the lasing line in the range 80°K-300°K which is in good agreement with the experiment.

I. INTRODUCTION

The origin of the lasing transition in ultra-pure GaAs is still an unsolved problem. Contrary to heavily doped P-N junctions in which the tail density of states introduced by doping accounts for most of the characteristics of the device,¹⁻⁴ in ultra-pure GaAs different mechanisms must be considered in order to account for the experimental observations.

At low temperature, theoretical calculations^{5,6} and experimental investigations⁷ suggest that an excitonic transition is responsible for the laser action. As the temperature increases, however, this laser action should be dominated by a direct band-to-band recombination process.⁸ At liquid nitrogen temperature, various investigations⁹⁻¹¹ initiated by Basov and co-workers, support well a model of band-to-band recombination but suggest that it must include many-body interactions in order to explain the experimental value of the lasing frequency. A careful study at 77°K¹² has shown that the electron-electron interaction¹³ mainly accounts for the experimental observations. At room temperature the lasing frequency appears to have an even stronger shift and, in order to have a complete understanding of the mechanism responsible for the transition, two independent studies of its temperature dependence have been reported.^{14,15} The results are controversial.

First Kressel and Lockwood reported observation of the lasing transition in the temperature range 4.2°K-345°K for p⁺-n-n⁺ double heterojunction (AlGa) As-GaAs laser. Because of the close confinement produced by the refractive index discontinuities, the observed radiation

is assumed to come only from the lightly doped n type region. In this case, they show that the lasing frequency does not follow the temperature shift of the band gap determined by Sturge.¹⁶ The discrepancy which is an increasing function of temperature varies from about 2 meV at 4.2°K up to 30 meV at room temperature. They concluded that even in their lightly doped samples ($n \sim 10^{16} \text{ cm}^{-3}$) the stimulated emission is not a simple band-to-band process.

Second, Chinn-Rossi and Wolfe¹⁵ reported some time later opposite results. Their work differs with the previous one in that both higher purity laser material (GaAs, $\text{Na+Nd} \sim 10^{14} \text{ cm}^{-3}$) and an optical excitation method were used. These authors measured both lasing energy and photoconductivity. At low temperature, they confirmed that the lasing frequency is well below the band gap but now, as the temperature increases, the difference between the lasing frequency and the band gap decreases. At room temperature, the laser frequency and the band gap determined from photoconductivity both converge to 1.407 eV. This result would, therefore, support the idea of a simple band-to-band recombination.

Such a conclusion of course depends on our knowledge of the band gap vs temperature. In Table I the value $E_0 = 1.407 \text{ eV}$ obtained at room temperature in the work of Ref. 15 is compared with different results previously published. We notice that the difference is quite large (about 65 meV). This leads to difficulty in commenting whether the lasing frequency agrees with the gap energy. Clearly both simple absorption¹⁶⁻¹⁸ and photoconductivity measurements¹⁵ are not sufficiently accurate. At room temperature the band edge becomes less abrupt, one must increase the resolution by performing a derivative of the spectra.¹⁹⁻²² Even then

there are difficulties in analyzing the data. For example, Pond and Handler²¹ have been able to fit the same electroreflectance spectrum of pure GaAs at 273°K with two different values of the transition edge depending on the theoretical model used: $E_0 = 1.427$ eV with a one electron theory of Franz-Keldish effect, $E_0 = 1.445$ with an excitonic theory of electroreflectivity.

To avoid such difficulties, we have measured both electroreflectance (E.R.) and piezoreflectance (P.R.) of GaAs with high resolution. We report the results in Section II. In Section III we compare the temperature dependence of the band edge with that of the lasing frequency and show that the difference is an increasing function of temperature. Finally, in Section IV, we discuss a simple model of band-to-band recombination which includes many-body interaction and show that it accounts well for most of the experimental results.

II. EXPERIMENTS

A schematic diagram of our experimental apparatus is given in Ref. 23. By simply changing the P.Z.T. transducers into an electrolytic cell, we are able to perform both P.R. and E.R. measurements. In both cases, the samples used were mechanically polished and chemically etched by standard techniques and the detector used was an S-20 photomultiplier.

A. Piezorefectivity Data

A typical P.R. spectrum obtained on high purity GaAs samples ($n = 1.5 \times 10^{15} \text{ cm}^{-3}$) is shown in Fig. 1A. It is compared with the theoretical derivative spectrum calculated²⁴ from Sturge's absorption data¹⁶ in Fig. 1B. Both spectrum are consistent with a very small perturbation of the band structure. The calculation²⁴ corresponds to $\Delta E_g = 3.10^{-4} \text{ eV}$. With a standard value of 50 kg/cm^2 for the A.C. stress applied on the sample and the hydrostatic pressure coefficient given for the fundamental gap of GaAs given by Paul and co-workers²⁵, we find: $\Delta E_g \sim 5.10^{-4} \text{ eV}$. This amplitude permits us to analyze our data in terms of a first derivative of the joint density of states and to achieve an accurate determination of the absorption edge.

Let us consider as an example the excitonic absorption curve (a 2-dimensional M_0 critical point) which corresponds to the step function of Fig. 2. In this case, the first derivative ΔE_2 is the resonant δ function given in Fig. 2A, if we assume a finite value for the broadening parameter Γ . We can show that $\Delta R/R$ is proportional to the real part $\Delta \epsilon_1$ of the modulated dielectric constant, given in curve 2-B, so the position of the excitonic absorption edge corresponds to the interception with the base line. The broadening parameter Γ is

easily obtained from half the energy difference between the two extrema.

At room temperature, from the spectrum given in Fig. 1A, we obtain:

$$E_0 = 1.424 \pm 0.002 \text{ eV and } \Gamma < 10 \text{ meV.}$$

Adding to this value an exciton energy of 3.8 meV^{22} and comparing this result to the values given in Table I, we find that our value of the band gap in GaAs at room temperature is:

- 7 meV lower than the value given by Sturge¹⁶
- 2 meV higher than the photoconductivity value of Ref. 15
- and lastly, 4 meV higher than the recent data of Sell and co-workers.²²

The main reason for this discrepancy seems to be in the lower purity of our samples ($n \sim 10^{15} \text{ cm}^{-3}$) as compared with the samples of Ref. 22 (vapor phase epitaxy $n \sim 5 \cdot 10^{13} \text{ cm}^{-3}$). Indeed, with decreasing purity, the band edge in GaAs is less sharp and slightly shifts to higher energy.²²

B. Electroreflectivity Data

A confirmation of our determination at room temperature is obtained by E.R. measurements on both n and p type GaAs at 293°K. A characteristic spectrum obtained on a p-type sample ($3 \cdot 10^{14} \text{ cm}^{-3}$) is given on Fig. 3. In the case of E.R., the line shape observed strongly depends on the magnitude of the electric field and the determination of material parameters from an experimental spectrum is difficult. However, for sufficiently low values of the modulating field, the experimental analysis is drastically simplified²⁶: a simple measurement of the asymmetry ratio of both extrema permit an independent determination of the absorption edge and of the broadening parameter Γ .

In the case of our experimental spectrum of Fig. 3, the modulation amplitude was only 50 meV. We have verified that this is in the low field limit by noticing, for example, that the signal amplitude is directly proportional to the square of the modulation field. Under these low field conditions the 3 points fit analysis of Aspnes²⁶ works well and one obtains at 293°K:

$$E_{\text{ex}} = 1.426 \pm 0.004 \text{ eV and } \Gamma = 20 \text{ meV}$$

These results are in good agreement with our P.R. determination and confirm that the lasing energy independently measured by Chinn, Rossi and Wolfe (1.407 eV) or Kressel and Lockwood (1.401 eV) in high purity GaAs are both far below the one electron band gap at room temperature. We study in the next section the temperature dependence of this difference between the lasing energy and the gap energy.

III. TEMPERATURE DEPENDENCE OF THE ONE ELECTRON BAND GAP

Figure 4 summarizes the temperature dependence obtained from our P.R. data and compares with other results in the same temperature range.^{16,27} They agree well, and give a value:

$$dE_0/dT = -3.9 \cdot 10^{-4} \text{ eV/}^\circ\text{K}$$

for the temperature coefficient of the fundamental edge between 100 and 300°K.

Figure 4 also shows the temperature dependence of the lasing energy obtained in Refs. 14 and 15. In the temperature range 77°K-300°K the two works agree well, they are also in agreement with the value given in Ref. 12 at 77°K. In the low temperature range, 2°K-60°K, there is a discrepancy between the lasing measurements on double heterojunctions and those on optically pumped GaAs.¹⁵ This can be accounted for by the presence of a small Al content in the active region of the double heterojunction diode²⁸ which slightly increases the band gap (~5 meV).

Figure 4 shows that even in pure GaAs the lasing energy is never the same as the band gap energy. In fact the temperature coefficient of the lasing energy between 300 and 100°K is $dE_L/dT \approx -4.5 \times 10^{-4} \text{ eV/}^\circ\text{K}$. This value is about 20% higher than the temperature coefficient of the fundamental edge and, therefore, the lasing energy appears to shift farther away from the band edge with increasing temperature: $(E_0 - h\nu_L)$ varies from 7 meV at 77°K to 20 meV at 300°K. Considering the differences

in samples purity, samples geometry and injection process in different works (optical pumping¹⁵ or electron beam excitation¹² of single platelets with an impurity concentration $7 \times 10^{13} < Na+Nd < 5 \times 10^{14} \text{ cm}^{-3}$, electrical injection of carriers¹⁴ in double heterojunction diodes with $n \sim 2 \times 10^{16} \text{ cm}^{-3}$ or electron beam excitation of the n type region alone²⁸) one should conclude that the good agreement among the various experimental results is an indication that the lasing energy is a characteristic property of the GaAs crystal itself.

First we must discuss two possibilities including residual impurities. The simple one is that lasing comes from carrier recombination at residual acceptor states. This can be easily dismissed. First the well known acceptor levels in GaAs have binding energy²⁹ of the order of 30 meV, well above the red shift of the lasing energy. Second the matrix element of the transition should be strongly dependent of the concentration. The standard relation:³⁰

$$T_{CA} \approx 10^{-17} N_A T_{CV}$$

where T is the transition probability, the subscript C, V and A refer to conduction band, valence band and acceptor states respectively and N_A is the acceptor concentration, shows that T_{CA} begins to be important only when $N_A \geq 10^{17} \text{ cm}^{-3}$. It remains almost negligible with $N_A \sim 10^{14} \text{ cm}^{-3}$. Lastly, the spontaneous emission from the conduction band to acceptor states has been previously observed on intentionally doped p type samples¹⁴ with $N_A \sim 10^{17} \text{ cm}^{-3}$ and in the temperature range of interest, its peak always shifts parallel to the band gap energy.

The possibility of a recombination at donor states is less easily dismissed. In all the zincblende semiconductors, the valence band undergoes a nearly rigid shift vs temperature:³¹ it moves without important deformation, so the ionization energies of acceptor states are independent of temperature. In opposite the conduction band shifts differently at points Γ , X and L. This gives rise to three different temperature coefficients for the interband transitions E_0 , E_1 and E_2 (at center and edges of the Brillouin Zone in [111] and [100] direction, respectively). These differences in temperature coefficients reflects the deformation of the band structure and are quantitatively related to the electronic wave functions at these different points.³¹ In the same way a transition between a donor level and the valence band will have a temperature coefficient which depends of the wave function which describes the impurity. For hydrogenic impurities, the wave function is a pure combination of Γ states and the temperature coefficient is the same as for the lowest gap: their ionization energy is independent of temperature. For deeper donor levels the wave function is no longer a pure combination of Γ states but includes more and more contribution of X and L states³² according to the energy differences Γ -L and Γ -X. For sufficiently deep donor levels one can find²³ a temperature coefficient which is close to dE_1/dT or dE_2/dT . Such levels are said: associated with L or X minima of the conduction band. The same results appear under pressure. In GaAs the energy separation Γ -X is ≈ 0.48 eV, independent of temperature³¹, and the association of impurity levels with X_1 minima of the conduction band has been previously reported by Paul³² and Sladeck³³ in pressure experiments. However, in this case one finds from Table II that the

temperature dependence of the ionization energy (red shift of the lasing line) would correspond to an admixture of $|L_1\rangle$ with $|\Gamma_1\rangle$ wave functions in order to give the experimental value $dE_L/dT \approx -4.5 \times 10^{-4}$ eV/°K. This result is opposite to the expected admixture of $|X_1\rangle$ with $|\Gamma_1\rangle$ states. Moreover, as for residual acceptors, the matrix element of the transition should be strongly dependent of the concentration of impurities. This is not observed. And, lastly, this level should give a band impurity transition which is not observed in differential spectroscopy.

The only possibility which accounts well for the shift of the lasing frequency is a temperature dependence of the gap shrinkage due to a change in electron-electron interaction with change of temperature. This is shown in the next section.

IV. TEMPERATURE DEPENDENCE OF THE LASING LINE

The injection of carriers in undoped semiconductors is known to induce two opposite perturbations:

- a downward shift of the band gap (shrinkage) due to electron-electron interaction.

- an upward shift of the band gap mainly due to the filling of the conduction band (Burstein shift). Under injection it appears in the conduction band a distribution of carriers which may be described by a quasi-fermi level: F_n , with a similar quantity F_p for the valence band. When $F_n + F_p > E_g$, lasing condition can be achieved and stimulated emission can be observed in the range:

$$E_g < E < F_n + F_p \quad (1)$$

For a given injection (F_n and F_p) one finds a definite spectrum of emission gain $g(E)$ and the position of the lasing line corresponds to the maximum of this spectrum. This shows that the lasing energy should always be higher than the bandgap energy.

The injected carriers lower the band gap in proportion to their concentration. This situation is similar to the case of heavy doping³ but now we can neglect the screening of residual impurities and the corresponding small tail of density of states. The shift of band gap is given in¹² according to Wolff:¹³

$$\Delta E \text{ (eV)} = - \frac{e}{2\pi\epsilon_0\epsilon_r} \left(\frac{3}{\pi}\right)^{1/3} N^{1/3} \quad (2)$$

This equation is valid only if r_e , defined as the ratio of inter-carrier spacing to the electronic Bohr radius in the crystal, is $\gg 1$.

For GaAs this corresponds to a limiting concentration of $n \sim 10^{17} \text{ cm}^{-3}$. A similar but less important effect should also occur for the valence band. With the heavy hole mass of GaAs,³⁴ the condition $r_h > 1$ is never fulfilled in the range of injection studied and we can neglect this effect.

To compute the temperature dependence of the lasing energy in pure GaAs, we must therefore calculate:

a) the variation with temperature of the carrier concentration (injection) necessary to overpass the losses and to reach a total gain $g \geq 1$ in the cavity. We choose at threshold the experimental value: $g_{th} = 20 \text{ cm}^{-1}$, given in the work of Ref. 14.

b) the gap shrinkage $\Delta E_G(T)$ which corresponds to the above injected carrier concentration and gives:

$$E_G(T) = E_0(T) - \Delta E_G(T) \quad (3)$$

c) the lasing energy $E_L(T)$ which corresponds to $g_{th} = 20 \text{ cm}^{-1}$ and is relative to the gap energy $E_G(T)$.

Standard expressions for the gain spectrum of a high purity semiconductor, assuming a \vec{k} conservation law and a parabolic density of states, are given in Ref. 1. Most recently, Stern⁸ calculated theoretical curves of gain vs current density for undoped GaAs, assuming also a k selection rule but with a more realistic nonparabolic density of state following Kane's $\vec{k} \cdot \vec{p}$ model.³⁵ The main result of the calculation is to show that with the non-parabolic density of states, the threshold current densities are about 70% as large as the values calculated with a parabolic density of states. In order to account for this effect of non-parabolicity, we used in the parabolic model an average recombination

constant $\langle B \rangle = 5.85 \cdot 10^8 \text{ cm}^3/\text{sec}$ which is for GaAs 0.65 times the standard value calculated from the measured parameters. In the temperature range 80°K - 300°K this gives values of injection, gain and current densities in good agreement with the work of Ref. 8. A comparison of these values, calculated with various models, is given in Table III for a typical gain $g = 50 \text{ cm}^{-1}$. Figure 5 shows in the range $10 < g < 200 \text{ cm}^{-1}$ the gain-current relations obtained in our calculation compared with those calculated by the nonparabolic model. The two sets of curves are in fair agreement which justifies the approximation made.

In Fig. 6 we give in the same gain region $10 < g < 200 \text{ cm}^{-1}$, the dependence of gain vs injected carrier concentration at 80°K , 160°K and 300°K . This calculation assumes equal injection of electron and holes in the material. This corresponds to the standard assumption of identical lifetimes for both kind of carriers. With the low threshold value $g = 20 \text{ cm}^{-1}$ given in Ref. 13, we find an injection which is respectively: $n = p = 2.2 \cdot 10^{17} \text{ cm}^{-3}$ at 80°K , $6 \cdot 10^{17}$ at 160°K , and $1.5 \cdot 10^{18}$ at 300°K .

The diminution of bandgap ΔE_G is computed from Eq. (2) with the standard value of the dielectric constant in GaAs $\epsilon_r = 10.9$. It is shown by the dotted line in Fig. 7. It is an increasing function of temperature which varies from about 15 meV at 800°K to 30 meV at room temperature. Figure 7 also gives a set of curves corresponding to the lasing energy $E_L(T)$ obtained at various temperature from the position of the maximum gain with respect to $E_G(T)$. The results are given as a function of injection. Using the calculated values of $E_G(T)$,

Eqs. (2) and (3), we then obtain the lasing energies given in Table IV and in Fig. 4 in satisfactory agreement with experiment. We find for the temperature coefficient of the lasing line a value of $-4.3 \cdot 10^{-4}$ eV/°K as compared with the experimental value of $-4.5 \cdot 10^{-4}$.

From Fig. 5, we also find a $T^{3/2}$ dependence for the threshold current density needed to reach a constant gain. With the value $g = 20 \text{ cm}^{-1}$, we have $T = 740 \text{ A/cm}^2$ at 77°K, 200 A/cm^2 at 160°K and 500 A/cm^2 at 300°K. This $T^{3/2}$ temperature dependence is very close to the experimental observation¹⁴ of $J \sim T^{1.4}$ and renders a further support to this model.

In conclusion, we have found that the temperature dependence of the lasing line in undoped semiconductors is well understood by taking into account two competing mechanisms. The first one is an effect of exchange interaction of carriers which diminishes the gap of lasing materials as compared to the one-electron band gap. The gap shrinkage in an increasing function of temperature since increased carrier injection is needed to reach the lasing threshold at higher temperature. The second one is a combined effect of band filling by injected carriers and of the carriers in Fermi distributions. With a simple model for the density of states we have found that the second effect is always smaller than the first one. The shift of the laser frequency observed is, therefore, an increasing function of temperature. We have calculated a temperature dependence which is in good agreement with the experiment.

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Table I. Comparison of different results for the band gap of GaAs at room temperature.

E_0 (eV)	Techniques Used	References
1.38	absorption	17
1.435±0.003	absorption	16
1.435	absorption	18
1.420±0.005	photoreflectance	19
1.425±0.015	piezoreflectance	20
1.427 } 1.445 }	electroreflectance	21
1.407	photoconductivity	15
1.424±0.001	differential reflectivity	22

Table II. Average temperature shift between 80°K and 300°K of the lasing energy and of the E_0 , E_1 , E_2 interband transitions, in units of 10^{-4} eV/°K.
 a. after Ref. 14-15
 b. this work
 c. R.R.L. Zucca and Y.R. Shen, Phys. Rev. B1, 2668 (1970).

	$h\nu_L$	E_0	E_1	E_2
$\frac{dE}{dT}$	-4.5	-3.9	-5.3	-3.6
	a	b	c	c

Table III. Comparison of gain coefficient and nominal current density computed for the same injection of carriers, with:

- a. parabolic model, $B = 9 \cdot 10^8 \text{ cm}^3/\text{s}$
- b. nonparabolic model, Ref. 8
- c. parabolic model, $\langle B \rangle = 5.85 \cdot 10^8 \text{ cm}^3/\text{s}$.

T(°K)	n = p (10^{17} cm^{-3})	a		b		c	
		g (cm^{-1})	I (A/cm^2)	g (cm^{-1})	I (A/cm^2)	g (cm^{-1})	I (A/cm^2)
80	2.7	78	1350	50	900	50	878
160	7.2	90	3600	50	2300	58	2340
300	18.0	112	9000	50	5300	70	5830

Table IV. Details of the contributions which determine the energy of the lasing line in pure GaAs (the value of E_0 include a constant excitation energy $E_{ex} = 4$ meV).

T (°K)	E_0 (eV)	g_{th} (cm^{-1})	n = p ($10^{17} cm^{-3}$)	ΔE_G (10^{-3} eV)	E_G (eV)	$h\nu_L - E_G$ (10^{-3} eV)	$h\nu_L$ (eV)
80	1.511	20	2.2	15	1.496	1	1.497
160	1.482	20	6	22	1.460	2	1.462
300	1.428	20	15	30	1.398	4	1.402

Figure Captions

Fig. 1. A) Piezoreflectance spectrum obtained on GaAs at room temperature.

B) Theoretical spectrum obtained by KRAMER-KRONIG transform of the absorption data (after ref. 24).

Fig. 2. Upper corner: Imaginary part of the dielectric constant ϵ_2 for a typical excitonic absorption edge.

A) Derivative $\Delta\epsilon_2$ of the preceding curve assuming a finite value Γ of the broadening parameter.

B) Modulation $\Delta\epsilon_1$ of the real part of the dielectric constant ($\Delta R/R$). This dispersion curve is simply the well known KRAMER-KRONIG transform of a resonant δ function. (For example see P. BATZ, Semiconductors and Semimetals 9, 315 (1972).)

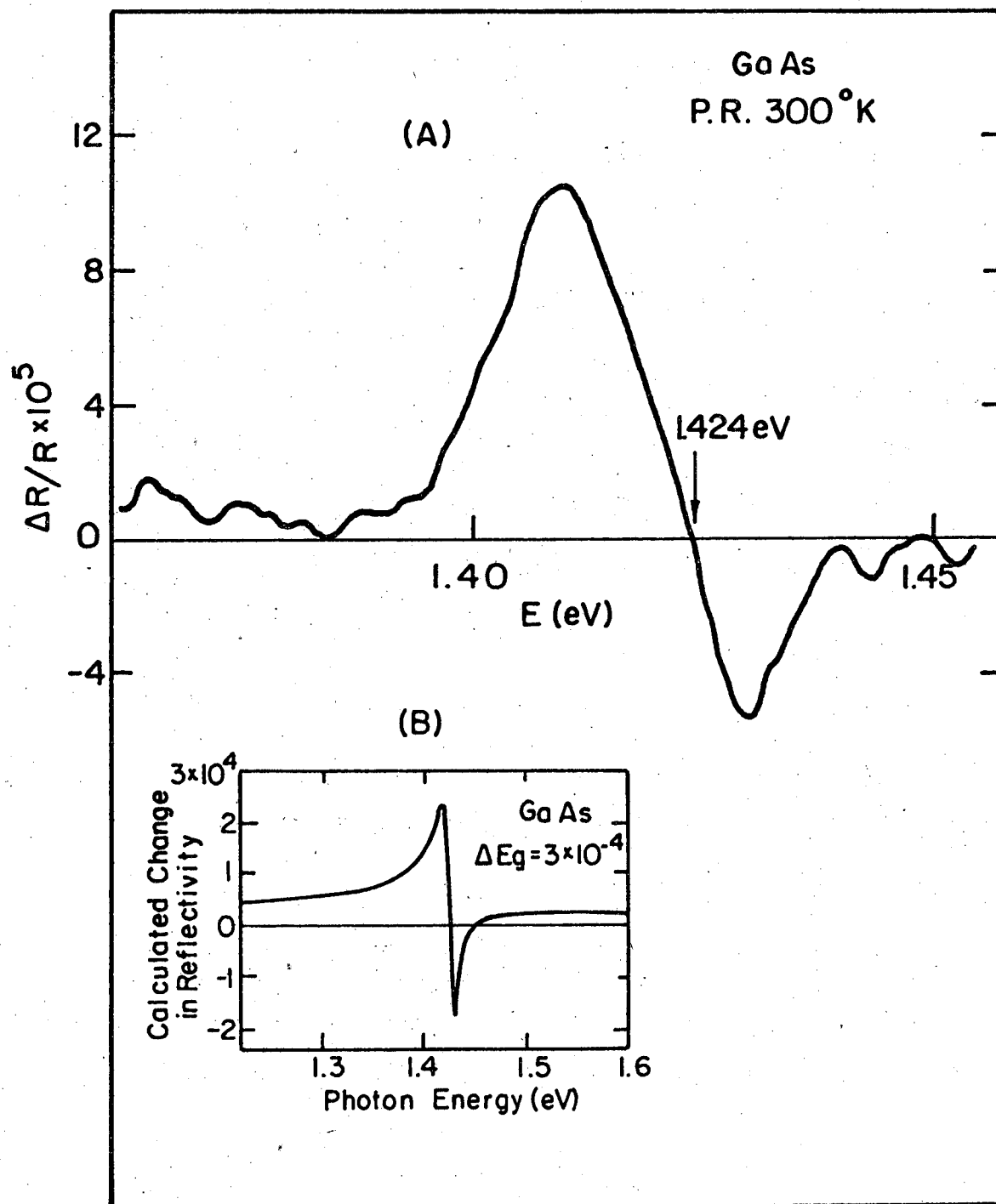
Fig. 3. Electro-reflectance spectrum obtained at room temperature on p type GaAs (Modulation: 50 meV).

Fig. 4. Temperature dependence of the one electron Band gap: x This work, \square Ref. 27, \circ Ref. 16. Also given in this figure are the experimental results obtained for the temperature dependence of the lasing line: \blacksquare ref. 14, \bullet ref. 15. The theoretical curve corresponds to the results of Section IV.

Fig. 5. Comparison of gain coefficient versus nominal current density obtained with: -- a parabolic model, this work
 - a non parabolic density of states, Ref. 8.

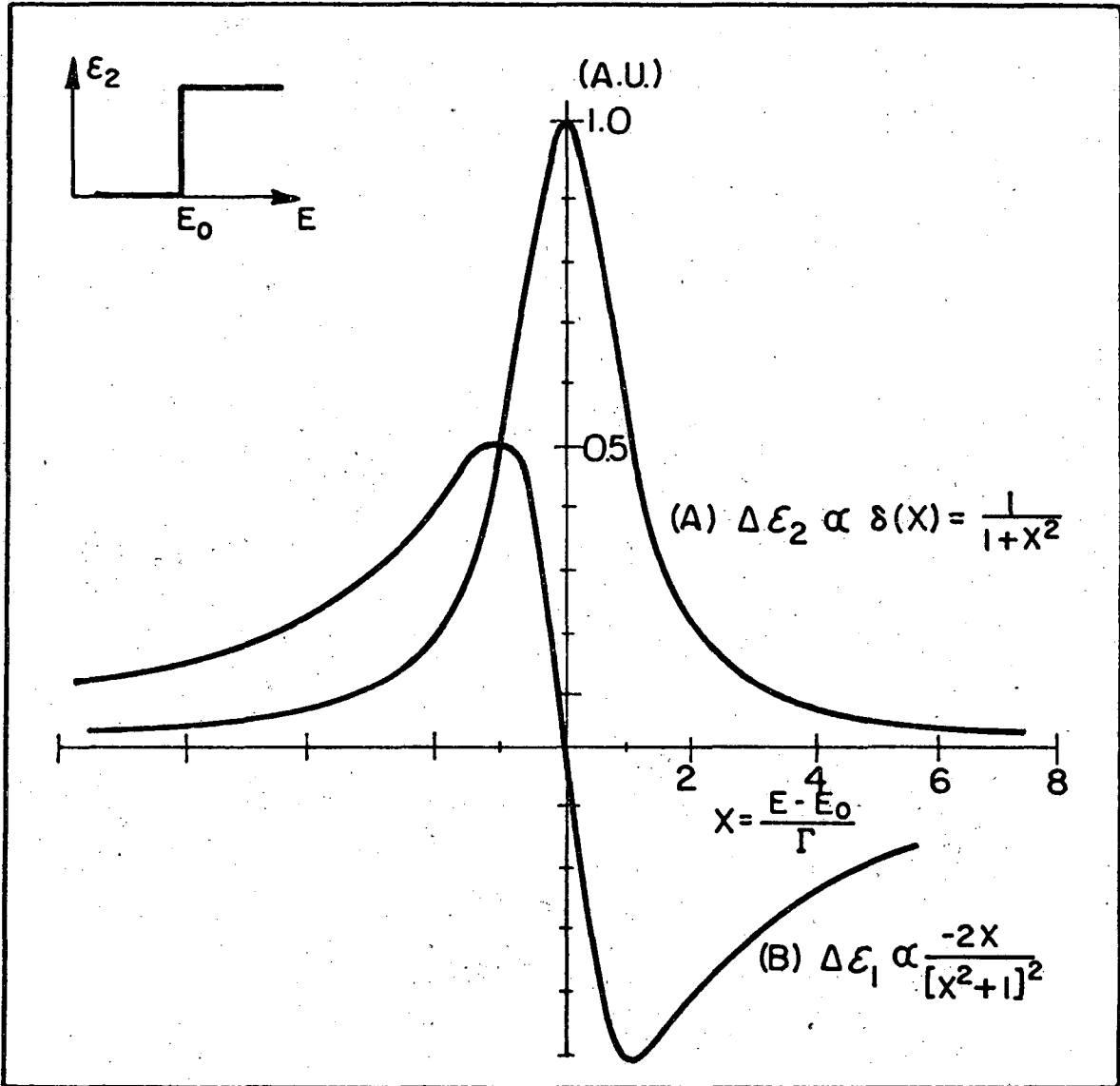
Fig. 6. Dependence of the gain coefficient versus injection for undoped GaAs. The points \blacksquare correspond to $g = 50 \text{ cm}^{-1}$ in the work of Ref. 8.

Fig. 7. Position of the maximum of $g(E)$ versus excitation for undoped GaAs. Dotted line, plot of the band shrinkage versus injection obtained from relation (2).



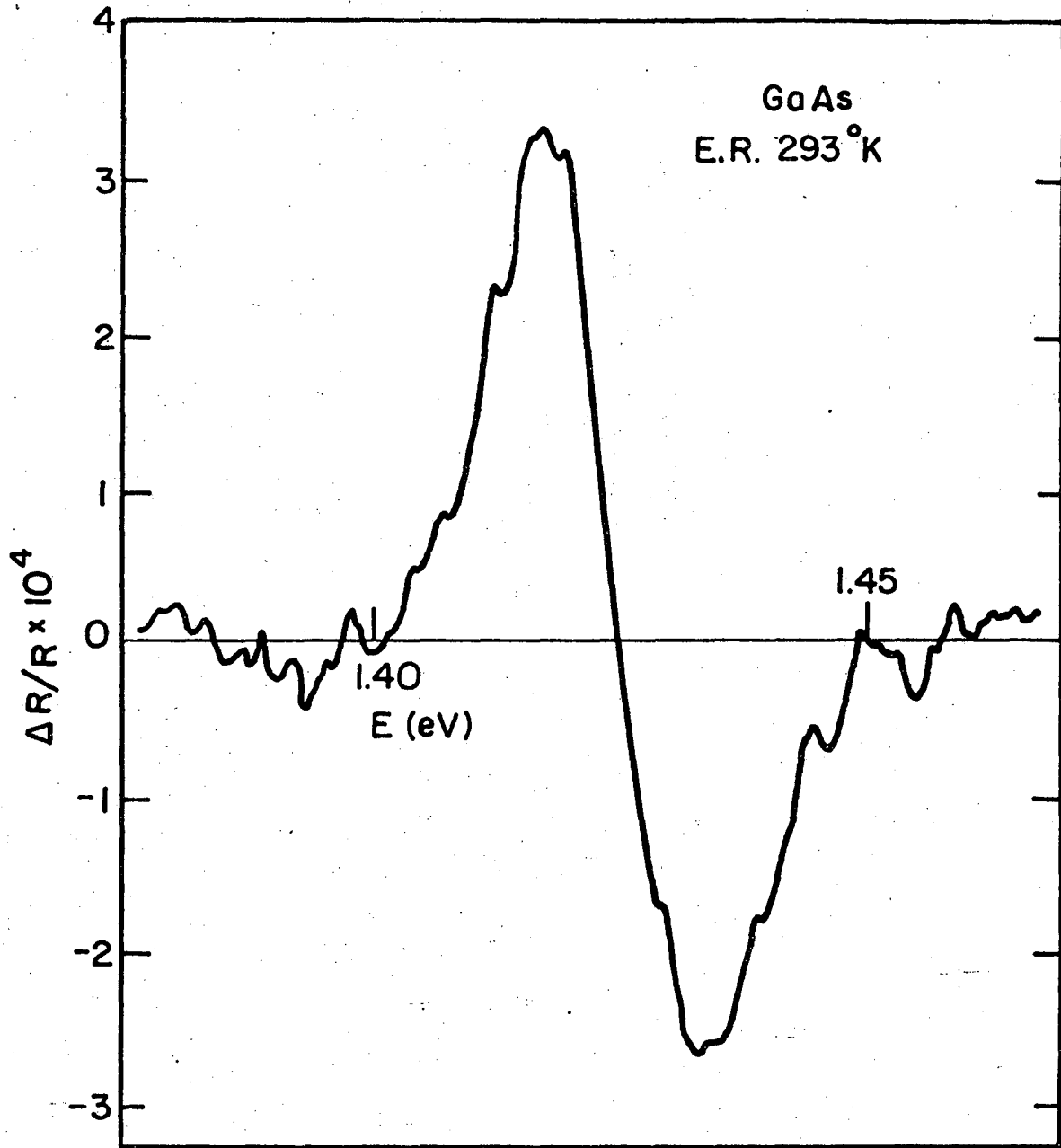
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Fig. 1



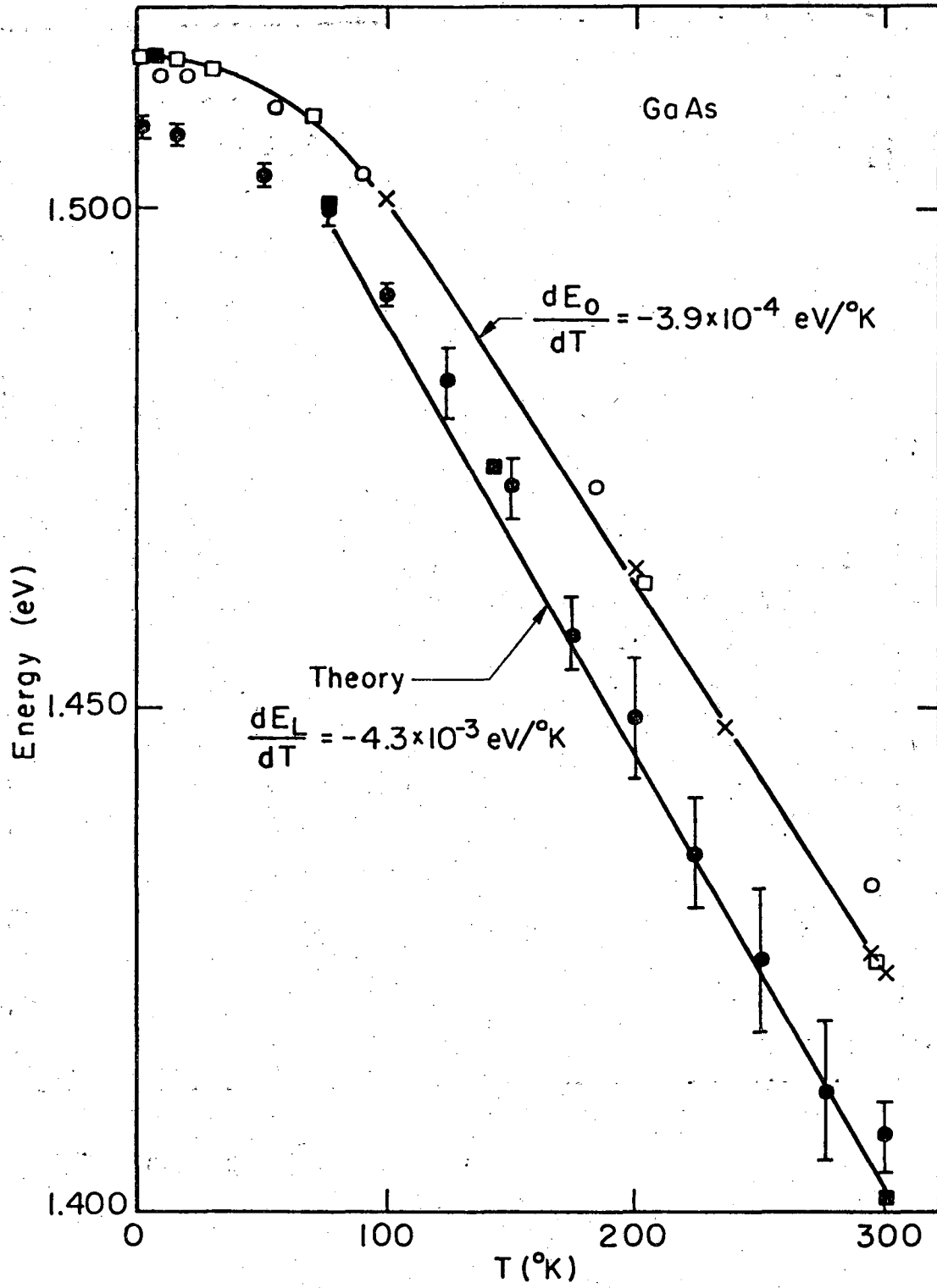
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Fig. 2



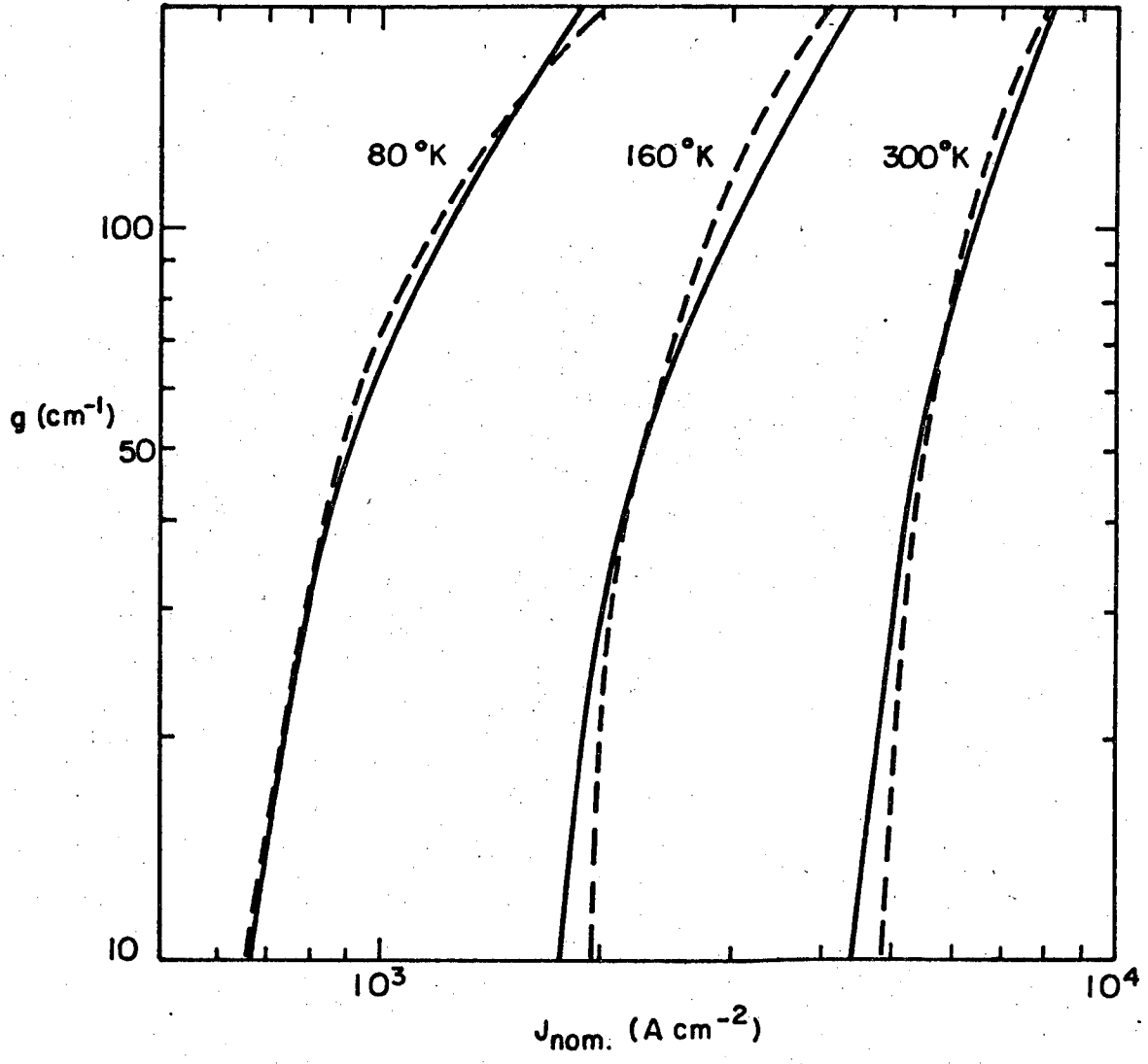
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Fig. 3



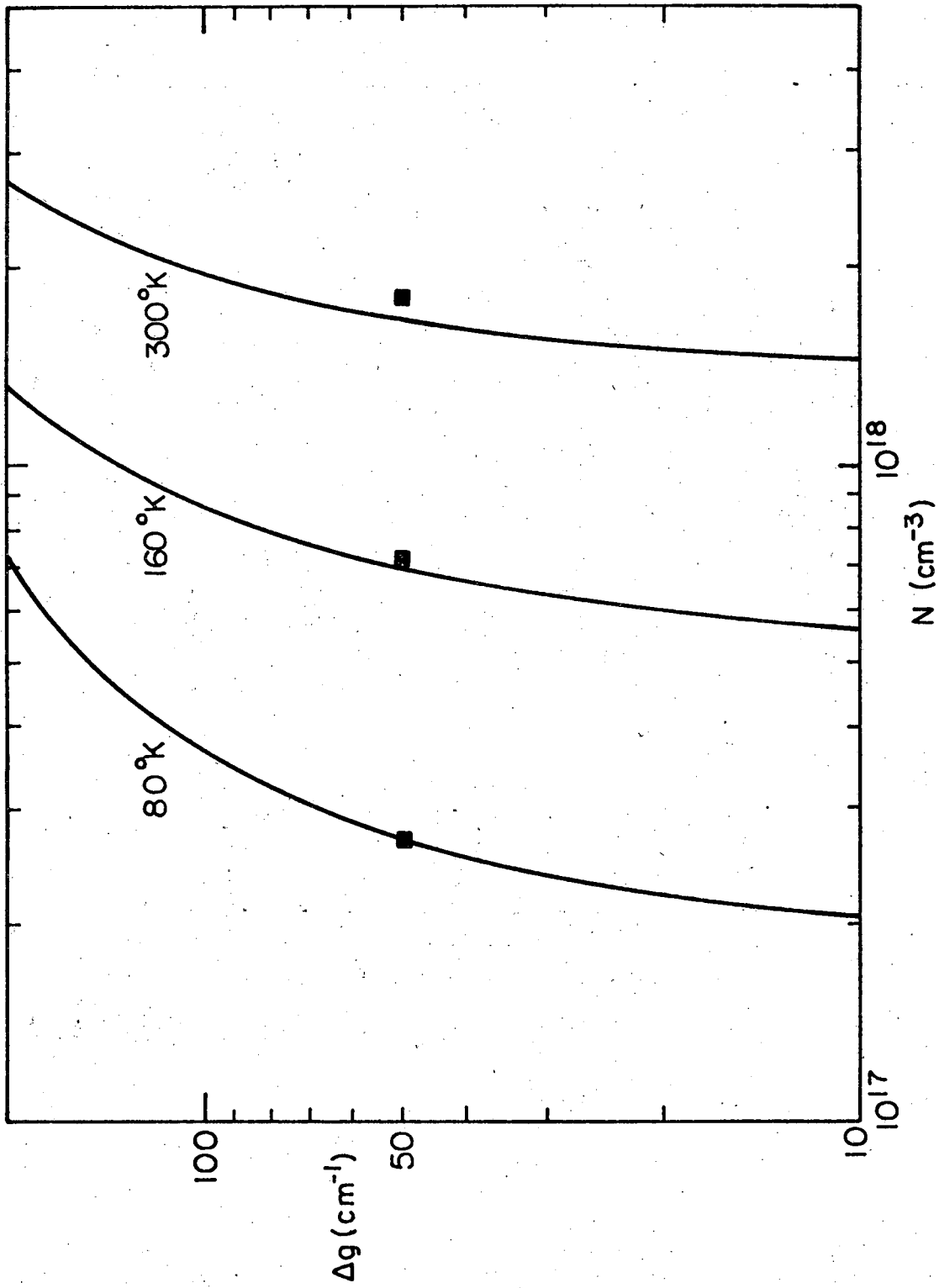
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Fig. 4



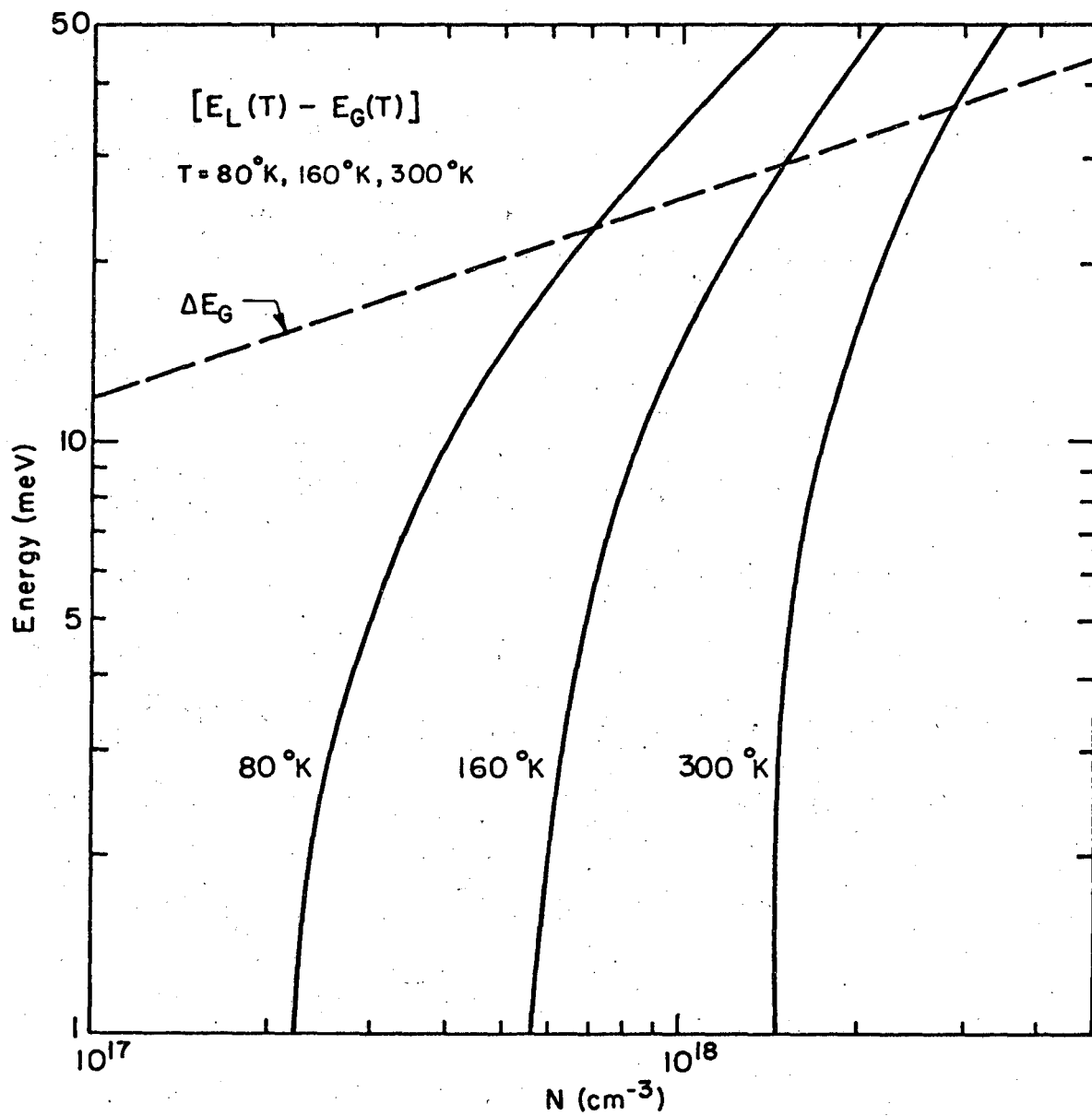
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• Fig. 5



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Fig. 6



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Fig. 7



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