Temperature dependence of GaAs random laser characteristics

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We show the temperature-dependent random lasing characteristics of photoexcited GaAs powders from 30 to 300 K. The lasing properties strongly depend on the temperature, i.e., the lasing peak energy increases and the threshold excitation power decreases as the temperature decreases. A theoretical model, in which the gain spectra of heavily doped *n*-GaAs are taken into consideration, well describes the temperature dependence of the lasing peak energy. The temperature dependence of the threshold excitation power can be also explained by the model qualitatively.

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I. INTRODUCTION

GaAs is a compound semiconductor material used for an injection-type semiconductor laser because of its high radiative electron-hole recombination rate due to the direct bandgap nature. Traditional semiconductor lasers have well-defined cavities, such as Fabry-Perot, distributed-feedback, and distributed-Bragg reflector resonators. Recently, stimulated emission sources without having any precise external cavities are demonstrated, called random lasers.[1,2] Random laser is first proposed by Letokhov in the late 1960s,[3] and is extensively studied using materials, such as ZnO (Ref. 4) and GaAs semiconductor powders,[5] ground solid-state laser materials,[1] and liquid dye with glass particles.[6] The random lasers consist only of irregularly shaped particles with a few hundreds nanometer sizes and the laser actions are thought to be caused by strong multiple scattering of emitted lights within these particles. Because of its simplicity and low cost, random laser is attractive candidate for use in new type of light-emitting source.

The lasing properties of random laser, such as lasing peak energy and the threshold excitation power, could be mainly determined by two factors. One is the scattering parameter. The threshold power and slope efficiency of lasing strongly depend on the size of the particles consisting of the random laser[7] and on the excitation pulse duration.[8] By controlling the Mie resonance of scatterers the peak energy of the random lasing can be modified[9] and resultantly the threshold power can be reduced.[10] Furthermore, the temperature tunability in the random laser was demonstrated using the liquid crystal having temperature-dependent scattering properties.[11] The second factor is the gain properties of an amplified medium. Yan *et al.*[12] showed that the change in the composition of a semiconductor alloy can shift the lasing peak energy and excitation threshold power. This is known to simply arise from the modification of lasing gain properties. These authors also demonstrated that the lasing properties are strongly dependent on the operation temperature.[12,13] This dependence is to be due to the temperature-dependent gain properties of the semiconductor laser material; however, the temperature dependence of such gain properties are not fully understood at present. It is interesting and important to know the temperature dependence of the lasing properties of random lasers. Because the gain of the semiconductor materials varies with temperature in a complicated manner, the prediction of the lasing properties is not so easy.

In this work, we investigate the temperature dependence of random laser characteristics of GaAs powders. The temperature-dependent lasing properties of GaAs random laser are compared to the theoretically calculated gain properties. GaAs is used as a typical laser material and its gain properties are well studied theoretically.[14] We demonstrate that the lasing energy position of GaAs random laser is strongly dependent on the temperature and that the dependence of the lasing peak energy is well explained by the temperature dependence of gain spectra of GaAs obtained from a theoretical model, which is popularly used in the traditional GaAs laser analysis. We also show that the temperature dependence of the threshold excitation power is qualitatively explained by the present model.

II. EXPERIMENTAL PROCEDURE

Commercially available *n*-GaAs single crystals (Si-doped, $n=1\times10^{18}-4\times10^{18}$ cm^{-3}) were ground in an agate mortar and dispersed in methanol. To obtain powdered GaAs film, the methanol solution was dropped on a Si substrate and then dried in room ambient. The size of each GaAs powder is several micrometers. Radiated lights from such particle films are strongly scattered that enable light amplification when optical gain exceeds absorption loss by high-intensity excitation. The powdered GaAs film was pumped by frequency-doubled light pulses (532 nm) from a neodymium-doped yttrium aluminum garnet (Nd:YAG) laser with 5 ns pulse duration or continuously. The pumping light was incident at an angle of 45° with respect to the sample surface. The size of the excited light spot was 1 mm^2 at the sample surface. The emitted light from the sample was collected in the directions of 90° from the sample surface. Photoexcited emission spectra were then measured using a single monochrometer equipped with a charge-coupled device (PIXIS:100B, Princeton Instruments). All optical measurements were performed in a closed-cycle refrigerator cryostat (IWATANI CRT 105PL) at 30–300 K.

III. EXPERIMENTAL RESULTS

Figure 1(a) shows the evolution of photoexcited emission spectra of powdered GaAs film at 300 K. At lower excitation

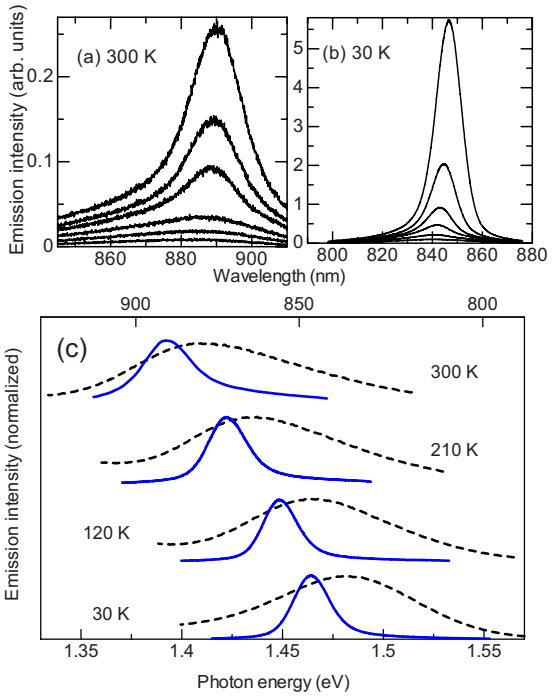

FIG. 1. (Color online) Evolution of photoexcited emission spectra from GaAs powders at (a) 300 and (b) 30 K and at excitation power densities from 0.9 to 5.4 MW/cm^2. (c) Spontaneous (dashed lines) and stimulated emission spectra (solid lines) from GaAs powders at 300, 210, 120, and 30 K.

power, the broad emission band at around 890 nm is observed. This band corresponds to the spontaneous emission of GaAs. Increasing the excitation power, the emission intensity steeply increases and simultaneously the linewidth decreases. Moreover, emission peak slightly shifts toward longer wavelength side. At 30 K [Fig. 1(b)], the essentially same behaviors are observed; however, each emission intensity is much larger than that measured at 300 K. The spectral evolution in Fig. 1 suggests that the stimulated emission occurs at above any threshold intensity. We note that the stimulated emission is never observed under continuous wave (cw) excitation. Figure 1(c) shows the comparison between spontaneous emission spectra excited by cw Nd:YAG laser and stimulated emission spectra taken at 300, 210, 120, and 30 K. The stimulated emission peak appears at the lower energy side of spontaneous emission band at all temperatures investigated, and the peak energies of spontaneous and stimulated emission increase with decreasing temperature. The origin of the temperature dependence of the emission peak energy will be discussed later.

In Fig. 2, emission peak intensity of GaAs powders is plotted as a function of the excitation power density at various temperatures. The emission intensity versus excitation power curves are strongly dependent on the temperature. The curves have two characteristic regions; one is the region where the spontaneous emission gradually increases with increasing excitation power and the other region is that the emission intensity steeply increases at above excitation threshold. The inset of Fig. 2 shows the full widths at half maxima (FWHMs) of the emission peak as a function of the

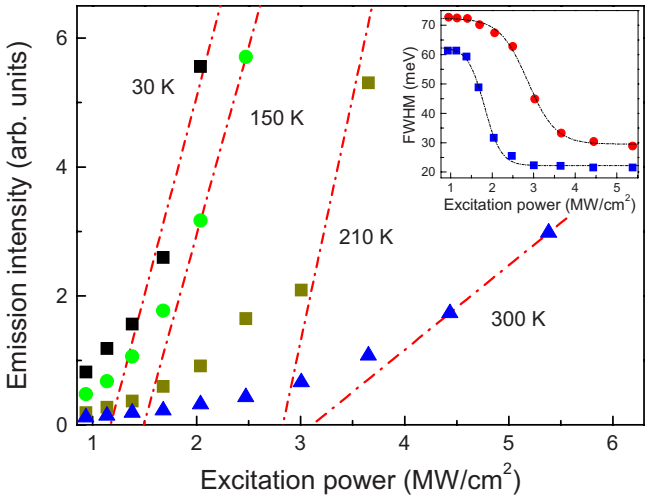

FIG. 2. (Color online) Emission intensity as a function of excitation power density at 300, 210, 150, and 30 K. The inset shows FWHM vs excitation power density plots at 300 and 30 K. Dashed lines are guide to eyes.

excitation power density at 300 and 30 K. The FWHM value steeply decreases at above threshold excitation power density.

The intensity and spectral narrowing in Fig. 2 indicate that random lasing in GaAs powders occurs due to the multiple scattering of emitted lights. In random lasers based on ZnO powders, several discrete narrow emission peaks due to the coherent (resonant) feedback of lights were observed on broad spontaneous emission spectra.[4] In our samples, no such peaks are found. One reason for this may be due to that our GaAs random laser is in the nonresonant feedback regime, in which the interference of light waves can be neglected and the motion of photons in a scattering medium can be treated as a diffusionlike manner, as similar to the case of solid-state laser materials.[1] Another reason is that the longer excitation pulses may average over each lasing shot and thus smear such peaks, although the transition from the incoherent to the coherent regime occurs as reported in $GaAs/Al_2O_3$ (Ref. 5) and polymer random lasers.[15]

We can estimate the threshold excitation power from Fig. 2. The estimated value of the threshold power density at 30 K is about 1.2 MW/cm^2. The order of this value is the same as that of the ZnO random laser.[4] The threshold is also found to be dependent on the temperature, e.g., 1.5 MW/cm^2 at 150 K, 2.8 MW/cm^2 at 210 K, and 3.1 MW/cm^2 at 300 K. The temperature dependence of the threshold will be discussed later.

IV. THEORETICAL MODEL

As shown in Fig. 1, the energies of lasing peak of GaAs powders are strongly dependent on the temperature. The lasing properties of ordinary laser can be determined from the two independent factors; the gain medium and optical resonator. In particular, the lasing energy can be easily tuned by the resonance frequency of the resonator. However, the random lasers do not have any fixed geometry resonator but the

feedback of light is caused by scattering in randomly shaped medium. This randomness results in the nearly constant scattering coefficient in the gain spectral band. Therefore, the lasing energy in random laser is expected to be determined from the maximum in the gain spectrum of the amplified medium. To explain the temperature dependence of the lasing energy in GaAs random laser, we calculate the gain spectra in GaAs at various temperatures. The theoretical model and procedure used in the calculation are as follows.

The gain spectrum $g(E)$ corresponds to the negative absorption coefficient $\alpha(E)$ in any absorption region, i.e., $g(E)=-\alpha(E)$. Calculation of the optical absorption in heavily doped semiconductors has been performed by several authors.[14,16,17] Let us use a model developed by Stern *et al.*[14,16,18] In this model, the optical absorption involved with transition between the valence and conduction bands in a one-electron model can be written as

$$-g(E)=\alpha(E)=\frac{\pi e^2\hbar}{\varepsilon m^2 cnE}\int_{-\infty}^{\infty}\rho_c(E')\rho_v(E'')|M_{cv}(E',E'')|^2 \times[f_v(E'')-f_c(E')]dE', \tag{1}$$

where m is the free-electron mass, ε is the permittivity of medium, E is the photon energy, $E''=E'-E$, n is the refractive index for E, ρ_c and ρ_v are the densities of states per unit volume and unit energy in the conduction and valence bands, respectively, f_c and f_v are the probabilities of that the states E' and E'' are occupied by an electron, respectively, and M_{cv} is the effective matrix element between the E' conduction-band and E'' valence-band states. In our calculation, the contributions of the heavy-hole and light-hole bands are taken into consideration.

Because our sample is the n-type degenerate semiconductor, the concentration-dependent density of states should be used. Thus, we have used the Gaussian-Halperin Lax band-tail model,[14,18] which gives the following conduction- and valence-band densities of states,

$$\rho_c(E')=(2\eta_c)^{1/2}(m_n^{3/2}\pi^2\hbar^3)y[(E'-E_c)/\eta_c], \tag{2}$$

$$\rho_v(E'')=(2\eta_v)^{1/2}(m_p^{3/2}\pi^2\hbar^3)y[(E_v-E'')/\eta_v], \tag{3}$$

where m_n (m_p) is the density of state mass for the conduction (valence) band, E_c (E_v) is the energy of the conduction-band (valence-band) edge, and the dimensionless variable y is defined in Ref. 19. The band-tail energy for the conduction (valence) band η_c (η_v) in Eq. (2) [Eq. (3)] is only parameter, which can be estimated from the Halperin-Lax band-tail density of states.[14,20] In the calculation of the Halperin-Lax density of states, we self-consistently determined the screening length L from the relation[17]

$$1/L^2=4\pi e^2/kT\varepsilon\left[\int\rho_c f_c(1-f_c)dE'+\int\rho_v f_v(1-f_v)dE''\right], \tag{4}$$

where k is the Boltzmann constant and T is the absolute temperature. The screening length in Eq. (4) depends on T and ρ_c (ρ_v). The occupation probabilities f_c and f_v are obtained from the Fermi-Dirac distribution function

$$f_c=(\exp[(E'-F_c)/kT])^{-1}, \tag{5}$$

$$f_v=(\exp[(E''-F_v)/kT])^{-1}, \tag{6}$$

where F_c (F_v) represents the quasi-Fermi level for the conduction (valence) band at nonequilibrium. The quasi-Fermi levels can be determined from the following expression,[14]

$$n=\int\rho_c(E-E_c)f_c dE, \tag{7}$$

$$p=\int\rho_v(E_v-E)(1-f_v)dE, \tag{8}$$

where n and p are the electron and hole concentrations, respectively. Because the quasi-Fermi levels are not influenced by the carrier-concentration-dependent band tail,[14] the parabolic form of the density of states is assumed in the calculation of the quasi-Fermi levels. The energy-dependent matrix element can be given by a product of the average matrix element M_b and the envelope matrix element M_{env},[21]

$$|M_{cv}|^2=|M_b|^2|M_{env}|^2. \tag{9}$$

For III-V semiconductors, M_b can be given by

$$|M_b|^2=m^2E_g(E_g+\Delta_{so})/12m_n\left(E_g+\frac{2}{3}\Delta_{so}\right), \tag{10}$$

where E_g is the energy gap and Δ_{so} is the spin-orbit splitting energy. The envelope matrix element M_{env} given by Stern[21] is used in the calculation.

The numeric parameters used here are $\varepsilon=\varepsilon_r\varepsilon_0=13\varepsilon_0$, $m_n=0.067m$, $m_{hh}=0.46$, and $m_{lh}=0.082$, where m_{hh} and m_{lh} are the heavy-hole and light-hole masses, respectively, and ε_0 is the permittivity in vacuum. The temperature-dependent intrinsic energy gap for GaAs E_g^0 is given by[22] (E_g^0 in electron volt, T in kelvin)

$$E_g^0(T)=1.517-5.5\times10^4T^2/(T+225), \tag{11}$$

and the concentration-dependent energy-gap shrinkage ΔE_g is[14] (in electron volt)

$$\Delta E_g=1.6\times10^{-8}(p^{1/3}+n^{1/3}), \tag{12}$$

with n and p in cm^{-3}. For the determination of the majority carrier concentration n, we used the charge neutrality condition

$$n+N_A^-=p+N_D^+, \tag{13}$$

where N_A^- and N_D^+ are the ionized acceptor and donor concentrations, respectively. In the present calculation, we assumed that N_A^- and N_D^+ are temperature independent and are equal to the impurity concentrations N_A and N_D, respectively.

The impurity concentrations N_A and N_D are determined as follows. The equilibrium spontaneous-emission intensity $I(E)$ can be calculated from $\alpha(E)$,[14]

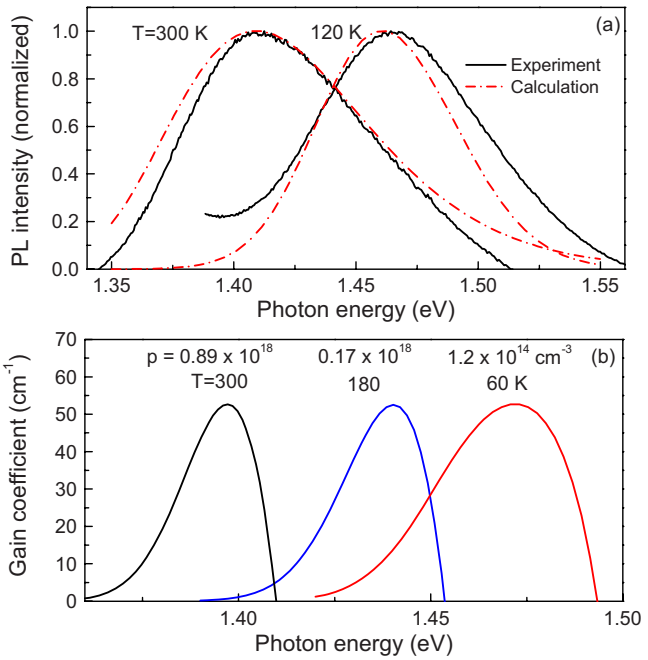

FIG. 3. (Color online) (a) Experimental (solid lines) and calculated spontaneous emission spectra (dashed lines) from GaAs powders at 300 and 120 K. The donor and accepter concentrations $N_D=1.5\times10^{18}$ cm^{-3} and $N_A=0.8\times10^{18}$ cm^{-3} are used in the calculation. (b) Calculated gain spectra at 300, 180, and 60 K. The minority carrier concentrations are $p=0.89\times10^{18}$ cm^{-3} at 300 K, 0.17×10^{18} cm^{-3} at 180 K, and 1.2×10^{14} cm^{-3} at 60 K. These values were estimated at the peak values of $g_{max}=50$ cm^{-1}.

$$I(E)=\frac{n^2E^2\alpha(E)}{\pi^2\hbar^3c^2[\exp(E/kT)-1]}. \quad (14)$$

The calculated $I(E)$ spectra are fitted to the experimental spontaneous-emission spectra shown in Fig. 1(c) with adjustment of N_A and N_D.

V. ANALYSIS AND DISCUSSION

Figure 3(a) shows the fitted results of the spontaneous emission spectra at $T=300$ and 120 K. The fit-determined N_A and N_D values are 0.8×10^{18} cm^{-3} and 1.5×10^{18} cm^{-3}, respectively. As shown in Fig. 3(a), the calculated spectra well reproduce the experimental spectral shape. The determined N_A and N_D values are, thus, considered to be reliable. In fact, the value of net impurity concentration ($N_D-N_A=0.7\times10^{18}$ cm^{-3}) is similar to the nominal Si impurity concentration of our sample ($n=1\times10^{18}-4\times10^{18}$ cm^{-3}). Note that the experimental spectra could not be reproduced without considering acceptor concentrations. Therefore, it is necessary to consider the presence of acceptors in the present analysis.

Using Eqs. (1)–(12), we can calculate the gain spectra at optional temperature T for minority carrier concentration p. Examples of our calculated gain spectra at $T=300$, 180, and 60 K are shown in Fig. 3(b). The minority carrier concentrations are determined at the maximum (peak) value of the gain coefficient $g_{max}=50$ cm^{-1}. The peak energy of gain spectrum shifts toward higher energy side with decreasing T, which is mainly due to the temperature shift of the band-gap energy [see Eq. (11)]. Moreover, the minority carrier concentration decreasing with decreasing T is the result of attaining the particular peak gain value. The decrease in p with decreasing T corresponds to the increase in the number of the excited carriers below the quasi-Fermi level.

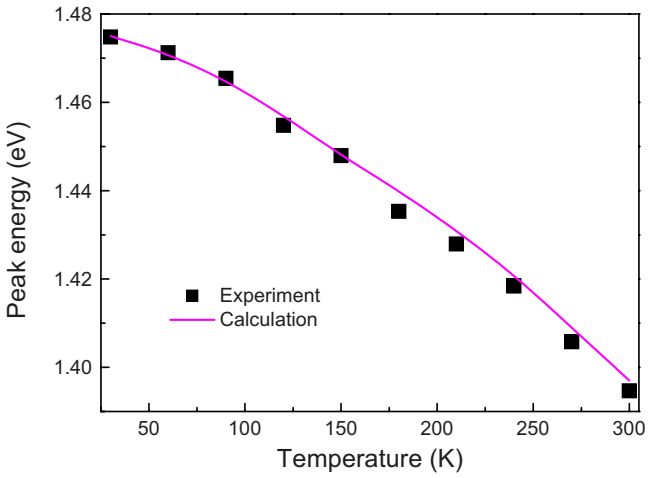

FIG. 4. (Color online) Temperature dependence of the lasing emission peak energy for GaAs random laser excited at the threshold power density (closed squares). Solid line represents the lasing energy calculated from the gain spectra.

Next, we compare the peak energies of our obtained gain spectra with those of the GaAs random laser measured at various temperatures. The lasing condition is that the gain must exceed any losses. In GaAs, the main losses should be the scattering and free-carrier absorption losses.[14] The required gain value for achieving lasing must be equal to or larger than the sum of these losses. Because the absorption loss is dependent of temperature, the required gain should also be dependent of it. In the present analysis, however, we assumed the losses to be independent of the temperature. Then, the required gain values can be regarded to be constant at all temperatures ($T=30-300$ K). The peak gain value ($g_{max}=50$ cm^{-1}, see below) is finally determined from our calculated gain curves with properly changing the minority carrier concentration over the temperature range $T=30-300$ K.

A comparison between the experimental lasing peaks and the peak energies of the calculated gain spectra at $g_{max}=50$ cm^{-1} is shown in Fig. 4. The excellent agreement between the experimental and calculated results is achieved over the entire temperature range. The fact suggests that the emission peak in the GaAs random laser can be uniquely determined at the maximum of the gain spectrum in bulk GaAs. It should be noted that the lasing peak energy is smaller than that of the intrinsic band-gap energy over the entire temperature range. This is to be due to the effect of band-gap shrinkage by the electron-electron interaction, which becomes more remarkable at higher minority carrier concentration p.

The experimental temperature dependence of the lasing energy in the conventional GaAs lasers was explained using

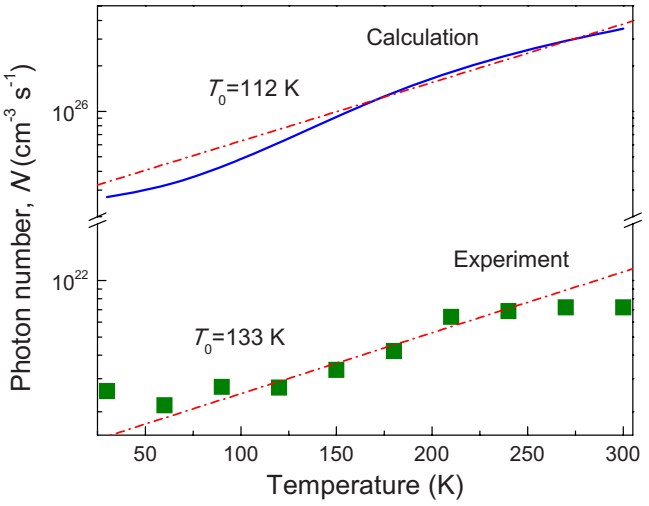

FIG. 5. (Color online) Temperature dependence of the threshold photon number per unit volume per second estimated from the experimental emission intensity vs excitation power density curves (solid squares) together with the calculation result (solid line). Dashed lines show fitted results with $N=N_0 \exp(T/T_0)$.

the essential same model as in the present study.[23] In that case, the gain spectra were calculated at $g_{\max}=20$ cm^{-1} which is much smaller than the present value. This indicates that GaAs random laser has larger losses, which is probably due to the stronger scattering and recurrent absorption events in the random medium than the conventional GaAs lasers.

As seen in Fig. 2, the threshold excitation power density of GaAs random laser depends on the temperature. To understand the temperature-dependent threshold, we estimate the threshold photon number from our obtained gain spectra $g(E)$. Assuming that the quantum efficiency is unity, the excited carriers radiatively recombine and emit spontaneous radiation below the threshold excitation. The photon number per unit volume per second N is given by[14]

$$N = \int_0^\infty r_{\text{spon}}(E)dE, \tag{15}$$

where $r_{\text{spon}}(E)$ is the spontaneous emission rate. The rate r_{spon} can be expressed in terms of the absorption coefficient[14]

$$r_{\text{spon}}(E) = \frac{n^2E^2\alpha(E)}{\pi^2\hbar^3c^2\{\exp[E-(F_c-F_v)]/kT-1\}}. \tag{16}$$

In the calculation of N, the calculated gain spectrum at $g_{\max}=50$ cm^{-1} is used.

Figure 5 shows the calculated photon number N as a function of temperature (solid line). The threshold photon number is estimated from the experimental values of the excitation threshold power density in Fig. 2 and is also plotted in Fig. 5 by solid squares. Here, the incident light is assumed to be fully absorbed by the sample. The temperature variation in the absorption coefficients at the excitation light wavelength is also corrected. As shown in Fig. 5, the experimentally obtained threshold photon number against T varies in the same manner as the calculated number. This result suggests that the temperature variation in the threshold is strongly correlated with the temperature-dependent gain properties. Note that the calculated photon numbers are much smaller than the experimentally obtained values at all temperatures ($T=30-300$ K). This is probably due to the higher quantum efficiency assumed in the calculation.

The fits to the threshold data with the empirical expression $N=N_0 \exp(T/T_0)$, where T_0 is the characteristic parameter, are shown in Fig. 5 (dashed lines). The experimental data in the $T=90-240$ K region are fitted because the data in this region have larger values than the experimental accuracy. The exponential function is popularly used to explain the temperature variation of the threshold current in the conventional lasers.[24] The experimental and theoretical T_0 values obtained here are 133 K and 112 K, respectively. The experimental T_0 value is almost in agreement with the theoretical value. We also note that the present T_0 values are comparable to those from the conventional GaAs injection laser[24] and also ZnO random laser.[13] We can see a difference in T_0 between the theoretical and experimental values. The reason for this may be due to no consideration of the temperature effect in the free-carrier absorption loss mechanism.

Noginov *et al.*[25] demonstrated that the threshold excitation power of GaAs/Al_2O_3 random laser can be well described by a model including the diffusive motion of photons. The threshold power in the incoherent regime of the random laser was considered to be proportional to the spontaneous emission rate and also $\sqrt{(l_t/l_{abs})}$, where l_t and l_{abs} are the transport mean-free pass and absorption length, respectively. l_{abs} is defined as the inverse of the absorption coefficient. Because l_{abs} depends on the temperature, developing a calculation model with including their model is still interesting work.

Broadening of the lasing spectrum at lower temperatures is observed in ZnO random laser with coherent feedback.[12] This may be interpreted by the broadening of the bandwidth of gain spectra [see Fig. 3(b)]. Therefore, it is expected that the lasing bandwidth of GaAs random laser with coherent feedback may broaden with decreasing temperature. To investigate the coherent regime of GaAs random laser is interesting and will be studied in the near future.

VI. CONCLUSIONS

We studied the temperature dependence of lasing properties of GaAs powders from 30 to 300 K. The lasing properties were strongly dependent on the temperature, i.e., the lasing peak energy increased and the threshold excitation decreased as the temperature decreased. Gain spectra of heavily doped n-GaAs at 30–300 K were calculated by considering the carrier concentration-dependent band tails and also energy-dependent momentum-matrix element. The calculated results well described the temperature-dependent lasing peak energy. It was concluded that the lasing peak energy in GaAs random laser can be determined from the maximum in the gain spectrum unlike the conventional lasers.[26] The temperature dependence of the threshold excitation, i.e., characteristic temperature, was qualitatively explained from the present results. This fact suggests that the temperature variation in the threshold is mainly determined by the

temperature-dependent gain properties. The present theoretical model can be further refined by including a diffusion theory or by considering temperature dependence of the absorption loss in GaAs. Such a refined model will predict exact lasing properties of semiconductor random lasers, including GaAs random laser.

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