Band-edge excitons in gallium arsenide on silicon

A. Freundlich, H. Kamada,* and G. Neu
Laboratoire de Physique du Solide et d'Energie Solaire, CNRS, Parc Sophia Antipolis, rue Bernard Gregory, 06560 Valbonne, France

B. Gil
Groupe d'Etude des Semiconducteurs, Université des Sciences et Techniques du Languedoc, place E. Battaillon, 34060 Montpellier CEDEX, France

(Received 29 September 1988; revised manuscript received 8 February 1989)

The electron light-hole and heavy-hole transitions in GaAs on Si are positively assigned by using low-temperature photoluminescence (PL) and reflectivity spectroscopies. The identification of the observed transitions was obtained from analysis of the oscillator strengths; then we could ascribe the high-energy transitions in the PL spectra to extrinsic recombination.

I. INTRODUCTION

An important issue in the field of semiconductor heteroepitaxial structures, such as GaAs directly grown on Si (GaAs/Si), is the existence of biaxial stress, which will alter the band structure, leading to a change in optical properties.

As has been well established from group-theory arguments, any biaxial stress in such materials will lift the degeneracy of the $J=\frac{3}{2}$ valence bands, leading to split subbands at $\mathbf{k}=0$ with different spin momenta $m_j=\pm\frac{1}{2}$ and $\pm\frac{3}{2}$.[1] Optical access such as low-temperature photoluminescence (PL) and photoluminescence-excitation (PLE) spectroscopies have revealed specific spectral features essentially due to these band splittings.[2–13] So far, the $m_j=\pm\frac{1}{2}$ and the $\pm\frac{3}{2}$ related recombination lines in the low-temperature (1.8–20 K) PL spectra in GaAs/Si are attributed to subband–to–conduction-band or intrinsic exciton transitions.[6–10] These experimental techniques, however, distinguish between different energy levels without direct information concerning the identity of observed energy states. In particular, it is not straightforward to distinguish an intrinsic exciton state from an extrinsic one or (and) the electron–light-hole state from the electron–heavy-hole one.

In this paper, combining PL and PLE spectroscopies with wavelength-modulated reflectance measurements (WMR), which are sensitive to the density of states as well as the selection rules, we identify undoubtedly the free exciton energies in GaAs/Si and confirm experimentally the association of $m_j=\pm\frac{1}{2}$ with the "heavy-hole" band and $m_j=\pm\frac{3}{2}$ with the "light-hole" band (in plane masses for normal optical access). Moreover, the observation of an interband transition in the 1.8-K reflectivity spectrum enables us to determine the exciton binding energy (rydberg) in GaAs/Si, in agreement with the theoretical approach for excitons in the strained GaAs crystal. Finally, the two highest-energy PL recombinations are found to be associated with extrinsic excitons (probably donor–bound-exciton recombinations).

II. EXPERIMENTAL DETAILS

In the following experiments, we have used 2.5-μm-thick metal-organic vapor-phase epitaxial (MOVPE) GaAs grown directly on (100) Si substrate tilted 2° toward [011]. Details of the growth process have been published elsewhere.[11,12,14] The layer was annealed *in situ* at 850 °C during the growth process, resulting in an improvement of both optical and structural properties.[12] The sample size used in present studies was 4×8 mm^2. The PL and PLE experiments were made using a 1-m-focal-length double-grating monochromator coupled with a photon-counting system and a dye laser pumped by a krypton-ion laser as an excitation source. The reflectivity experiments were made in a manner similar to that described previously.[15] The reflectivity spectrum was recorded with a lock-in amplifier, choosing a time constant of 1 sec. The wavelength modulation of the reflectivity was also recorded with a lock-in amplifier but with a time constant of 3 sec; the spectrometer was scanning with a speed of 50 Å/min. Experimental spectra being noisy, 24 of them were superimposed using a personal computer. The accuracy limit was as high as 0.3 meV due to the difficulty in extracting experimental features from a relatively high level of noise. All the experiments were performed in a geometry where the incident beam was always set perpendicular to the GaAs(100) epitaxial surface.

III. RESULTS AND DISCUSSIONS

The 2-K WMR and reflectivity spectra of GaAs/Si are presented, respectively, in Figs. 1(a) and 3. The WMR spectrum reveals one strong feature with a minimum peaking at 1.502 eV and a couple of weak features with minima peaking at 1.485 and 1.507 eV. Corresponding features are also visible in the spectrum without wavelength modulation (Fig. 3).

The 2-K PL spectra exhibit two high-energy features at 1.500_5 eV ($E_{\pm3/2}$) and 1.483_4 eV ($E_{\pm1/2}$) [Fig. 1(b)] and two low-lying acceptor-impurity related bands at 1.457

and 1.464 eV (not represented in Fig. 1) which were previously assigned as the $m_j=\pm\frac{1}{2}$ components of, respectively, Si and C donor-acceptor pair transitions.[12,13] The PLE spectra [Fig. 1(c)] exhibit two absorption thresholds with strong resonances at 1.502 and 1.485 eV, their linewidth being typically 1.8 meV. Note that these PLE resonance energies agree quite well with those of the reflectivity and WMR signals $X_{\rm hh}$ and $X_{\rm lh}$ presented above, inferring strong absorptions at these photon energies. Moreover, a clear energy correspondence between the $E_{\pm1/2}$ and $E_{\pm3/2}$ PL lines and the two sets of WMR structures can be manifested, though luminescence lines are slightly shifted toward lower energy.

A. Effects of the biaxial stress on the band structure of GaAs/Si: Identification of split free excitons

For GaAs grown on Si(100), the difference in the thermal expansion coefficients between GaAs and Si causes in the postgrowth cooling process a considerable amount of residual biaxial tensile stress (2–3 kbar at 2 K) in the (100) epitaxial planes.[4–13] Under (100) coplanar stress, the stress-tensor component along the [100] direction, chosen here as the quantization axis, is equal to zero. The system strain tensor is then given by $\epsilon_{xx}=\epsilon_{yy}=(S_{11}+S_{12})X$, $\epsilon_{zz}=2S_{12}X$, and $\epsilon_{xy}=\epsilon_{yz}=\epsilon_{zx}=0$, where S_{11} and S_{12} are the compliance coefficients, X is the stress magnitude, and x, y, and z refer, respectively, to the [001], [010], and [100] directions. According to the stress Hamiltonian of Bir and Pikus,[1] the strain tensor has a nonvanishing component of quadrupolar symmetry $2\epsilon_{zz}-\epsilon_{xx}-\epsilon_{yy}$ which will split the fourfold-degenerate $J=\frac{3}{2}$ valence band into two subbands with spin momentums of $m_j=\pm\frac{1}{2}$ and $m_j=\pm\frac{3}{2}$ by an energy difference of $E_S=2b(S_{11}-S_{12})X$, where b is the tetragonal shear deformation potential. The hydrostatic strain component ($\sim\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz}$), on the other hand, causes the shifts of both the Γ_6^c-like conduction band and $p_{3/2}$ multiplet center of gravity, i.e., a band shrinkage by $E_H=2a(S_{11}+2S_{12})X$, where a is the hydrostatic deformation potential. Accordingly, the energy shifts of the $m_j=\pm\frac{1}{2}$ and $m_j=\pm\frac{3}{2}$ valence subbands with respect to the Γ_6^c conduction band at $\mathbf{k}=\mathbf{0}$ are therefore described to first order by

$$\delta E_{g\pm1/2}=[2a(S_{11}+2S_{12})+b(S_{11}-S_{12})]X\ , \tag{1a}$$

$$\delta E_{g\pm3/2}=[2a(S_{11}+2S_{12})-b(S_{11}-S_{12})]X\ . \tag{1b}$$

Note that, considering the shear splitting only, the biaxial stress acts like a uniaxial one but with the sign changed. X is, in this notation, taken to be positive in the case of tensile stress.

To evaluate the stress magnitude, however, these equations cannot be directly applied to the split luminescence bands without knowing their exact characters.

Turning now to the experimental energy coincidences of the two reflectivity signals of 1.502 eV ($X_{\rm lh}$) and 1.485 eV ($X_{\rm hh}$) and PLE absorption resonances, we may undoubtedly associate these with the strong intrinsic optical transitions related to $m_j=\pm\frac{3}{2}$ and $m_j=\pm\frac{1}{2}$ subbands. Moreover, previous works[15,16] suggest identifying the minima of the structures at 1.502 eV ($X_{\rm lh}$) and 1.485 eV ($X_{\rm hh}$) in the WMR, on account of their line shape, as the split transverse free-exciton ground states ($1s$). It should be noticed that PLE resonances appear as positive contributions to the PL bands; once the free exciton is created, it is immediately trapped to slightly lower-energy states within its radiative lifetime. This is common for materials containing a rather high density of defects, such as impurities, as well as dislocations. The subsequent processes will be radiative recombination yielding the observed luminescence lines.

The slight differences between the exciton absorption energies and PL energies then suggest the extrinsic character of the $E_{\pm1/2}$ (1.4834 eV) and $E_{\pm3/2}$ (1.5005 eV) PL lines. Since the present material exhibits n-type conductivity, the neutral donor bound to excitons (D^0X) and/or neutral donor to band (D^0h) related recombinations are the primary candidates. This latest attribution is supported by the fact that the stress dependence of the donor state in GaAs is very weak (the localization energy of donor-bound excitons in GaAs is consistent with the

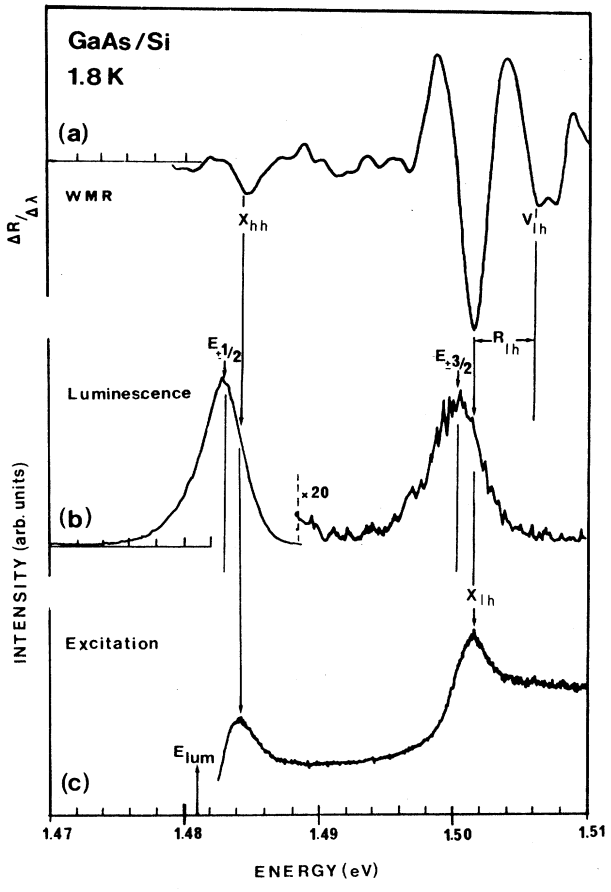

FIG. 1. 1.8-K spectra taken from 2.5-μm-thick GaAs on Si(100) of (a) wavelength-modulated reflectivity, (b) PL, and (c) PLE with $E_{\rm lum}=1.482$ eV. $R_{\rm lh}$ and $V_{\rm lh}$ are, respectively, the $m_j=\pm\frac{3}{2}$ exciton binding energy (Rydberg) and the $m_j=\pm\frac{3}{2}$ subband–to–conduction-band transition.

present observation), and also by the most recent work of Zemon *et al.*[10]

B. Heavy-hole and light-hole excitons

Under the biaxial stress the valence band decouples; the k-dependent parts of the valence-band energies are given in the lowest order in $\mathbf{k}$ in the $\mathbf{k}\cdot\mathbf{p}$ Hamiltonian as shown by Hensel and Feher,[17] and in atomic units ($\hbar=1$ and $m_0=\frac{1}{2}$) as

$$-\delta V_{\pm 1/2}=(\tau_1-\tau_2)k_\perp^2+(\tau_1+2\tau_2)k_\parallel^2 , \tag{2a}$$

$$-\delta V_{\pm 3/2}=(\tau_1+\tau_2)k_\perp^2+(\tau_1-2\tau_2)k_\parallel^2 , \tag{2b}$$

where τ_1,τ_2 are Luttinger parameters,[18] and where $k_\parallel=k_z$ and $k_\perp^2=k_x^2+k_y^2$. The constant-energy surfaces near $\mathbf{k}=\mathbf{0}$ are ellipsoids of revolution, whose principal axes are those of the reduced strain tensor. Split valence bands in biaxially strained GaAs are then schematically illustrated in Fig. 2(b). The light-hole and the heavy-hole masses are then anisotropic and $\mathbf{k}$-direction dependent. Since the optical access is commonly made normal to the GaAs(100) plane (light polarization is normal to the quantization axis z) as in the present experiments [Fig. 2(a)], it is a straightforward argument that optical excitation will involve in-plane masses. One will observe the "light-hole" and "heavy-hole" masses for the $m_j=\pm\frac{3}{2}$ and $m_j=\pm\frac{1}{2}$ bands, respectively.

Relative reflectivity signal amplitudes of the two-exciton band at 1.485 and 1.502 eV are the further experimental evidence of this assignment. The selection rules can be easily deduced for the transitions between the Γ_6^c conduction band and the split $m_j=\pm\frac{3}{2},\pm\frac{1}{2}$ subbands; with respect to the stress direction the excitation from the $m_j=\pm\frac{3}{2}$ band is allowed only in the polarization $E\perp X$. The exciton state will be roughly denoted, neglecting the electron-hole exchange interaction, as a system consisting of an electron and hole coupled via Coulomb interaction; thus its transition may be treated in a manner similar to the band-to-band transition. In the absence of stress, the oscillator strengths of the excitations relevant to $m_j=\pm\frac{3}{2},\pm\frac{1}{2}$ valence bands are in a ratio of $\frac{1}{2}$ and $\frac{1}{6}$ for the polarization $E\perp X$ (in the present experiment geometry).[19] This will be also the case when the splitting of the $J=\frac{3}{2}$ valence band E_S is enough smaller than the spin-orbit splitting Δ_0 (0.34 eV in GaAs). An estimate of the intensity for the $m_j=\pm\frac{3}{2}$ exciton relative to that for $m_j=\pm\frac{1}{2}$ can be obtained by calculating the ratio of interband matrix elements[20]

$$\tfrac{1}{2}/[\tfrac{1}{6}(1-2\delta E_S/\Delta_0)]=3.3 \tag{3}$$

times the ratio of joint densities of states[21]

$$\frac{(\tau_0+\tau_1-2\tau_2)^{-1/2}(\tau_0+\tau_1+\tau_2)^{-1}}{(\tau_0+\tau_1+2\tau_2)^{-1/2}(\tau_0+\tau_1-\tau_2)^{-1}}=1.01 \tag{4}$$

times the ratio of the oscillator strengths for $|\frac{3}{2},\frac{1}{2}\rangle$ and $|\frac{3}{2},\frac{1}{2}\rangle$ excitons.

To estimate the last quantity, we have for simplicity neglected any anisotropy of the static dielectric constant in strained GaAs; then we deduced the wave-function amplitude at the origin, $f(0)$, from the scaling relation between $f(0)$ and the effective Rydberg (R_0), well established for the $1s$ hydrogenic systems: $f(0)\propto(R_0)^{3/2}$. The effective Rydberg differs for $|\frac{3}{2},\frac{1}{2}\rangle$ and $|\frac{3}{2},\frac{3}{2}\rangle$ excitons due to the mass anisotropy. We expressed the Rydbergs according to with Bir and Pikus,[22]

$$R_i=R_0\left[1+\frac{\alpha_i^2}{10}\right] , \tag{5}$$

where $i=\{m_j=\pm\frac{1}{2}$ or $m_j=\pm\frac{3}{2}\}$, and R_0 is the bulk GaAs Rydberg. We have calculated the mass anisotropy factor $\alpha_i=3(m_\parallel-m_\perp)/(m_\parallel+2m_\perp)$ for the $m_j=\pm\frac{1}{2}$ and $m_j=\pm\frac{3}{2}$ bands leading to, respectively,

$$\alpha_{\pm 1/2}=\frac{-3\tau_2}{\tau_0+\tau_1+\tau_2} , \tag{6a}$$

$$\alpha_{\pm 3/2}=\frac{3\tau_2}{\tau_0+\tau_1-\tau_2} . \tag{6b}$$

Here τ_i ($i=1,2,3$) are the Luttinger parameters. Using the values of $\tau_0=(0.067)^{-1}$, $\tau_1=6.85$, and $\tau_2=2.1$ for GaAs,[18] it is rather straightforward to see that the two Rydbergs in the above first-order approximation are almost equivalent to that in unstressed GaAs. This calculation shows that exciton energies in strained GaAs shift with the same rates as the related subgaps.

The ratio of squared exciton wave-function amplitudes at the origin is given by

$$\left|\frac{f_{3/2}(0)}{f_{1/2}(0)}\right|^2=\left[\frac{10+[3\tau_2/(\tau_0+\tau_1-\tau_2)]^2}{10+[3\tau_2/(\tau_0+\tau_1+\tau_2)]^2}\right]^3=1.01 . \tag{7}$$

The above-considered contributions [Eq. (3)×Eq. (4)×Eq. (7)] lead to a calculated value of ~3.3 for the ratio of reflectivity structures, while the experimental intensity ratio in the reflectivity spectrum of Fig. 3 is around

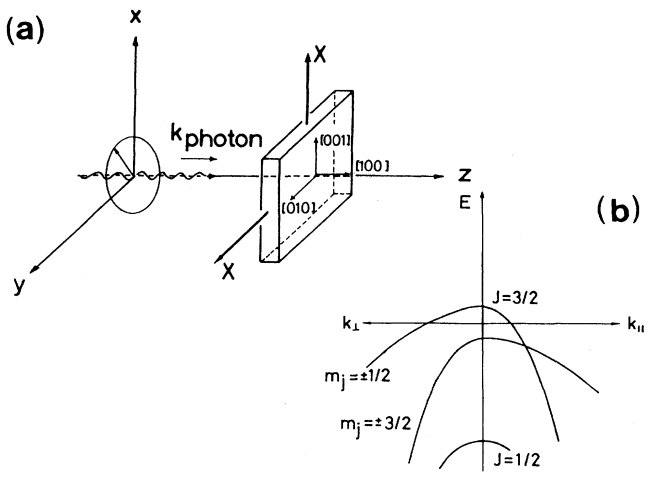

FIG. 2. Schematic representation of (a) the incident laser-beam polarization and the experimental geometry, and (b) the $\mathbf{k}$-dependent valence-band structures in biaxially strained GaAs.

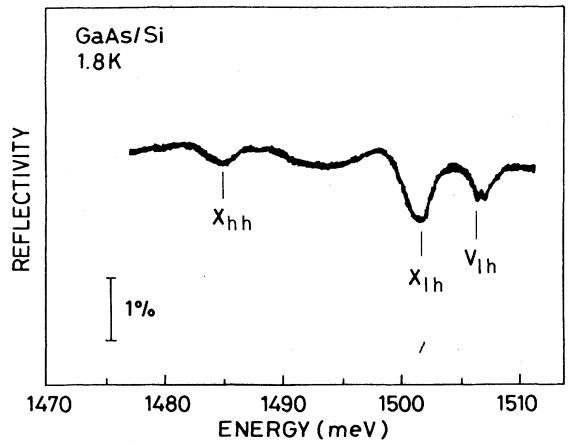

FIG. 3. 1.8-K reflectivity spectrum (without modulation) showing the X_{lh} intensity ratio of about 4 relative to that of X_{hh}.

4, somewhat larger than the above prediction. The slight disagreement may be due in part to the crudeness of our estimate of the wave-function amplitude at the origin for excitons in strained GaAs. Note also here that the magnitude of the reflectance structures is very small; the change in the reflectivity was only 0.9% of the total magnitude. Also, the great sensitivity of the $m_j=\pm\frac{1}{2}$ valence subband to the biaxial stress (because of its larger stress coefficient) will smoothen and diminish the structure in the possible case of inhomogeneity of the strain field in the layer. The agreement is, nevertheless, satisfactory.

C. The 1.507-eV structure

Now let us consider the large structure at ~ 1.507 eV. Recent work on PLE spectroscopy of GaAs on Si has made possible the observation of the $n=2$ excited state of the $m_j=\pm\frac{3}{2}$ related exciton.[9] However, the structure at 1.507 eV in our reflectivity spectrum cannot be associated only with the excited states of excitons since the intensity ratio between the structure at 1.507 eV and the exciton ground state at 1.502 eV in the reflectivity spectrum is larger than $\frac{1}{2}$. The oscillator strength for the (ns) excited state of the exciton is known to be smaller by a factor of $1/n^3$ than for the $(1s)$ ground state (e.g., the ratio should be in the range of $\frac{1}{8}$ for the $n=2$ excited state).[23]

Moreover, we believe that we can rule out the possibility of large strain fluctuations in the epilayer,[7,13] since the thickness of the GaAs epilayer in our sample is smaller than the critical thickness for the appearance of microrelaxation of strain in GaAs on Si, and spatial investigations of the luminescence spectra *never revealed* a luminescence band at 1.507 eV and a corresponding low-energy companion at higher energy than 1.485 eV. Furthermore, additional PLE spectroscopy measurements in the 1.485–1.502-eV range did not reveal any features other than those reported in Fig. 1(c).

To propose a plausible identification of the 1.507-eV reflectance structure we also have to consider the following points: (i) the shape of the structure at 1.507 eV in the WMR spectra is typical of the derivative of continuum-to-continuum transition [Fig. 1(a)],[24] and (ii) the energy separation R_{hh} between the minimum of the structure at 1.507 eV (V_{hh}) and the $m_j=\pm\frac{3}{2}$ exciton (X_{hh}) is about 4.8 ± 0.3 meV, which is consistent with the binding energy of the exciton ground states in bulk GaAs.

The change in the reflectivity near the excitonic resonances is very small compared with the change generally observed in high-quality GaAs (see, for instance, Fig. 1 of Ref. 25). We suggest that surface microfields ionize the exciton, and that, consequently, band-to-band processes have comparable intensity with excitonic ones and can be observed. Therefore, in view of the line shape, the intensity, and the energy position, the line at 1.507 eV (V_{hh}) is attributed to the interband transitions involving the $m_j=\pm\frac{3}{2}$ state of the valence band. Although the contribution from excited states in the low-energy tail of the structure at 1.507 eV might be possible, the main contributions should be due to interband transitions. The $m_j=\pm\frac{1}{2}$ subband–to–conduction-band transition intensity should be lower by at least a factor of 3 than the V_{lh} transition and its detection is probably shadowed by the relatively large background noise in the reflectivity spectra (Fig. 3).

Now the nature of the observed PL lines, their localization energies, and the Rydbergs are established. Hence we can estimate the stress magnitude in the material by taking them into account and applying Eqs. (1). Using the values published in the literature of $S_{11}=1.16\times10^{-6}$ bars^{-1}, $S_{12}=-0.37\times10^{-6}$ bars^{-1}, $a=8.41$ eV, and $b=1.76$ eV,[26] the stress coefficients of 4.37 meV/kbar and 9.76 meV/kbar for the $m_j=\pm\frac{3}{2}$ and $m_j=\pm\frac{1}{2}$ subbands are obtained. The stress value is estimated at 3.05 ± 0.05 kbar at 1.8 K for the sample used.

IV. CONCLUSION

Based on optical-selection-rule analysis we have for the first time experimentally determined the "electron–light-hole" and "electron–heavy-hole" transitions in GaAs on Si(100). It is cautioned that for the optical spectroscopy experiments, where usually the incident beam is normal to the epilayer surface, the $m_j=\pm\frac{3}{2}$ and $m_j=\pm\frac{1}{2}$ holes must refer to in-plane masses. Extension of such an experimental identification to the other covalent heterostructures (biaxially strained) is straightforward. Further, the observation of the $m_j=\pm\frac{3}{2}$ subband–to–conduction-band transitions at 2 K and the identification of both $m_j=\pm\frac{1}{2}$ and $m_j=\pm\frac{3}{2}$ free excitons based on WMR line-shape analysis allowed us to gain localization energies of the high-energy features in the PL spectra [tentatively attributed to donor-related recombinations (D^0h or D^0X)] and to estimate, for the first time, the exciton binding energy in GaAs/Si consistent with the theoretical prediction.

ACKNOWLEDGMENTS

The authors are grateful to C. Vèrié, A. C. Beye, M. Leroux, and A. A. M'Baye for fruitful discussions and to

A. Kozacki for his expert technical assistance. They are indebted to H. Mathieu of the Université des Sciences et Techniques du Languedoc (USTL), Montpellier for valuable suggestions. This work has been supported by the French Energy Management Agency (AFME), the European Community Contract, and Nippon Telegraph and Telephone Corporation (NTT) Optoelectronics Laboratories.

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*Also at Nippon Telegraph and Telephone Corporation (NTT) Optoelectronics Laboratory, 3-1, Morinosato Wakamiya, Atsugi, Kanagawa 243-01, Japan.

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