Reflectance spectroscopy on GaAs-$Ga_{0.5}Al_{0.5}As$ single quantum wells under in-plane uniaxial stress at liquid-helium temperature

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Reflectance spectroscopy is performed at 2 K on GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells by molecular-beam epitaxy, for different widths of the GaAs confining layer and when uniaxial stress is applied perpendicularly to the growth axis. The modifications of the electronic states produced by the stress are experimentally determined via changes of the reflectance-energy characteristics of light-hole excitons and heavy-hole excitons and theoretically accounted for by calculations in the framework of the envelope-function formalism.

I. INTRODUCTION

Uniaxial stress as an external perturbation has proved to be a powerful tool for studying the electronic states of crystals. It allowed valuable information to be obtained for both direct- and indirect-band-gap semiconductors.[1–3] Furthermore, it was used with success in the case of hydrogenic impurities,[4–6] isoelectronic bound excitons,[7] and biexcitons.[8] More recently, it was used to identify the symmetry of noncubic centers in silicon,[9,10] gallium phosphide,[11,12] etc. Coming now to the field of heterostructure physics, only a few contributions have been published from either the experimental side[13–15] or the theoretical one.[16–18] Some interesting findings are expected since it has been established that (i) the hydrostatic-pressure dependence of the excitonic features is a sensitive function of the confinement energies of the carriers,[19–22] and (ii) biaxial dilations, when lattice mismatch occurs between the confining material and the barrier, may have a notable influence on the relative ordering of electronic states.[23] In this paper, we first present an experimental investigation of the influence of an in-plane uniaxial stress on the characteristic transition energies of both heavy-hole and light-hole excitons confined in GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells for well widths ranging from 31 up to 124 Å. The experimental findings are then compared with the predictions of a theoretical calculation and a close agreement is found between the measurements and the theory.

II. EXPERIMENTAL METHODS

The samples investigated in this work were grown by molecular-beam epitaxy (MBE) in the Optoelectronic Joint Research Laboratory, Japan. The [001]-oriented GaAs substrate used in this study was cut within $\pm 0.1°$ out of a Cr-doped semi-insulating HB ingot. The preparation for MBE growth was as follows. The substrate was degreased in organic solvents rinsed in deionized water and etched in a 4:1:1 sulfuric acid:hydrogen, peroxide:deionized water solution for 2 min. After being rinsed in deionized water the substrate was dried in a stream of nitrogen gas. After this standard preparation process the substrate was mounted with molten indium on a molybdenum block. This block was loaded into the MBE growth chamber.

The sample structure consisted of (a) GaAs buffer layer (500 nm), (b) $Al_{0.5}Ga_{0.5}As$ buffer layer (300 nm), and (c) $Al_{0.5}Ga_{0.5}As$-GaAs single quantum wells (SQW's) with four different well widths separated by 50-nm-thick $Al_{0.5}Ga_{0.5}As$ barriers. The growth temperature was 600 °C and the As:Ga flux ratio was about 4. The growth rate of GaAs was about 0.5 monolayer/s. Growth was interrupted for 2 min at each heterointerface of SQW's in order to reduce interface-roughness-induced statistical broadening of optical line shapes.[24]

The external stress being applied in the [110] direction, the strain field and the [001]-oriented well potential are in crossed configuration. Reflectivity spectra were taken at pumped-liquid-helium temperature, and the focused broadband light from a tungsten-wire lamp was reflected from the sample onto the monochromator slit and subsequently dispersed and detected. Concerning the magnitude of the uniaxial stress, it could be measured from the mechanical deformation experienced by a quartz device located under the sample. It could also be deduced by studying the stress splitting of three-dimensional (3D) excitons freely propagating in the GaAs buffer. Magnitudes obtained with these mechanical and electronic gauges were found to be in close agreement.

III. EXPERIMENTAL FINDINGS

Figure 1 illustrates the stress patterns obtained for both GaAs and QW(1) ($L_z \sim 124$ Å). GaAs-related structures lie on the left-hand side of the figure. Looking at the higher-energy peak of GaAs, one observes for the high-energy level a stronger shift toward high energy than for the lower-energy one. This is due to competition between the hydrostatic and shear part of the strain.[1] No care has been taken to eliminate the influence of the grating on the polarized photons and some slight variance can be found here with Ref. 1 when considering the intensities of these structures. Let us now focus on the reflectance structures associated with excitons confined in QW(1). These transitions lie on the right-hand side of the figure. The lower-energy peak (the strongest one) can be interpreted as originating from heavy-hole excitons (HHE's), while, at zero stress, at higher energy, one recognizes a reflectance structure corresponding to light-hole excitons (LHE's). When the stress is increased, this latter state shifts faster than the former one does. Such a behavior will be discussed at length in the theoretical section. In the bulk case, it is well established that many physical quantities, such as refractive index, effective mass, and damping parameter, together with the so-called additional boundary conditions, contribute in a sensitive manner to the general shape of a reflectance structure.[2] The breaking of [001] translational symmetry in the case of QW's leads to supplementary complications for solving Maxwell's equations; a quantitative comprehension of these patterns in terms of the exciton-polariton formalism is outside the scope of the present paper. Note some weak modulations of the envelope of the reflectance structure. Arguments based on the effective-mass approximation suggest that they are interface-roughness effects. These interface-roughness effects, which cannot be well resolved for the wide structure QW(1), will be well resolved for QW(2) when the layer thicknesses is about 79 Å. As an illustration, Fig. 2 displays some stress patterns characteristic of QW(2). Careful examination of the reflectance structure corre-

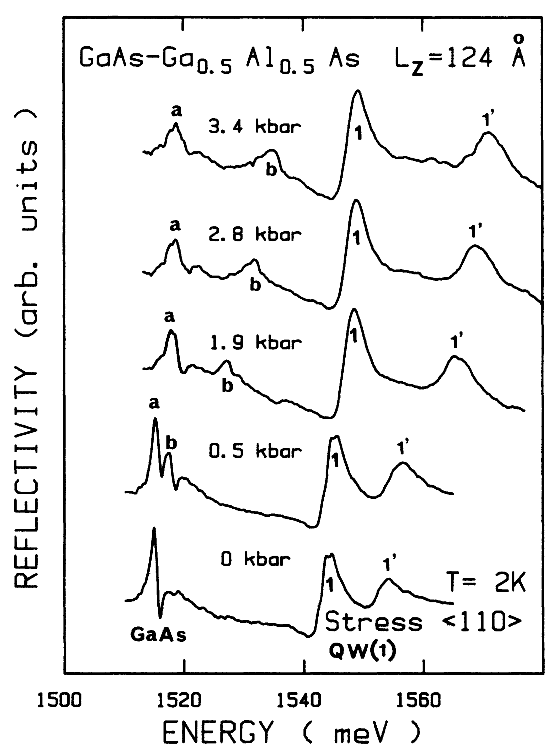

FIG. 1. Some typical reflectance spectra collected in the (1500–1580) meV range for stress between 0 and 3.4 kbar. To the left, the bulk-related structures, labeled *a* and *b* respectively, correspond to bulk-type light-hole and heavy-hole excitons (see Ref. 1). The density of states is heavier for *b* than for *a*, and the corresponding oscillator strengths should be measured stronger for *b* than for *a*. This is not found here because of the influence of the experimental setup (grating of the monochromator), which has not been corrected. To the right, we show the optical transitions corresponding to QW(1) ($L_z \sim 124$ Å). The unprimed structures correspond to heavy-hole excitons (HHE's), the primed ones correspond to light-hole excitons (LHE's). LHE's shift faster toward high energy than HHE's do. At low stress the envelope of the reflectance structure corresponding to HHE's displays some undulations which correspond to well-width fluctuations (see text).

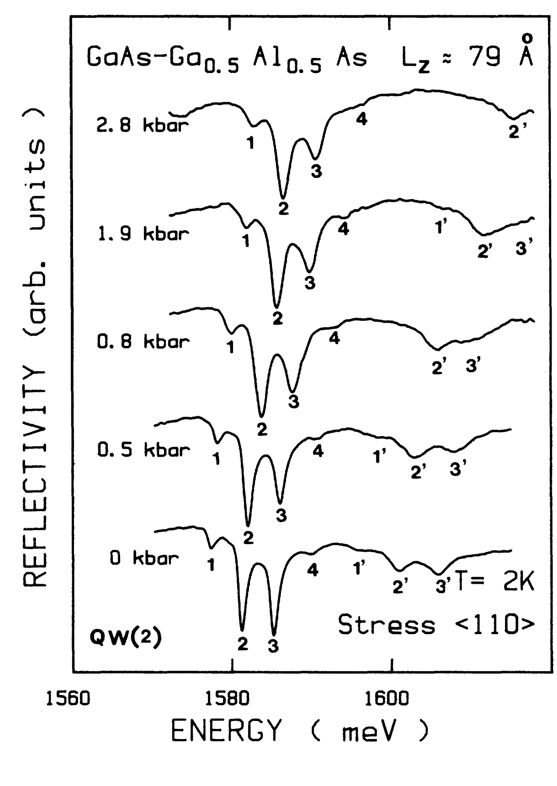

FIG. 2. The analog of Fig. 1, but for QW(2) ($L_z \sim 79$ Å). We clearly select four quantized levels for heavy-hole excitons labeled from 1 up to 4. Such spectra testify to the columnar structure of the SQW's grown with growth interruption at the heterointerfaces (see Ref. 24 in text). Primed transitions correspond to LHE's. In the high-stress range, the energy of the 3′ and 4′ states coincide with HHE's of QW(3) ($L_z \sim 53$ Å) and consequently can hardly be measured.

sponding to heavy-hole excitons reveals four well-resolved reflectance structures. In the case of growth interruption at the heterointerfaces, the reduction of the roughness of the quantum well has been optimized in such a way that interisland thermalization of carriers is widely suppressed and, instead of a wide single transitions, four transitions can be resolved; this is an evidence of the "terrace" structure of the interfaces.[24] The magnitude of each transition reflects the relative density of well-width fluctuations to which it corresponds. In the previously discussed case of QW(1), the energy separation between these transitions was too small to be resolved. The sharpness of the HHE transitions corresponding to QW(2) is retained when the stress is increased. In the case of the higher-energy levels (corresponding with the so-called light-hole excitons at zero stress), we again find four structures, but, in that case, due *partly* to a more important delocalization of the carriers in the barriers, the oscillator strengths are smaller than for HHE's. Shape criteria enable us to associate each low-energy transition with a higher-energy companion. Once more, the "LHE" transition shifts faster toward high energy than its lower-energy associate does. In order to limit the number of experimental patterns, we will not give the 53-Å [QW(3)] results [quite close to QW(2) in intensities], but we go directly to 34 Å [QW(4)] (see Fig. 3). In this case, the spread of the carriers into the barriers has been drastically enhanced by decreasing the well thickness, the structures broaden, and the oscillator strengths diminish. The splitting due to interfacial roughness is about 20 meV and an overlap of heavy-hole excitons and light-hole ones occur. However, one can unambiguously measure two HHE-type transitions (labeled) 1 and 2) and a light-hole-type one (labeled 2′). The situation is much more complicated in the 1750-meV region, where an overlap between 3 (HHE) and 1′ (LHE) strongly prevents an accurate determination of the transition energy at zero stress.

Furthermore, structures 3 and 1′ exhibit different stress coefficients, and in the range of stress available in this in-

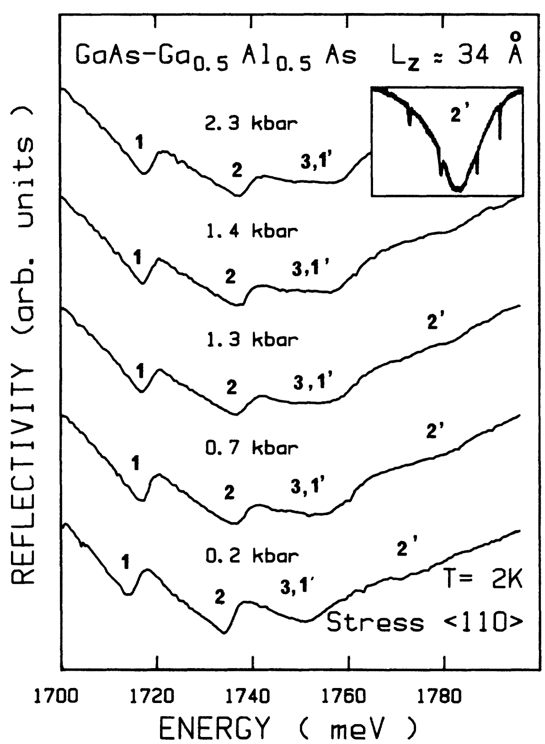

FIG. 3. In the case of QW(4) with $L_z \sim 34$ Å, the interface-roughness-induced splitting is comparable with the HHE-LHE splitting and the reflectance patterns are complicated to analyze. Levels labeled 1, 2, and 3, respectively, correspond to thicknesses of 34, 31, and 28 Å (typically) for the confining GaAs layer. States 1′ and 3 overlap. As for transitions 2′ (LHE) associated with 2 (HHE), they are well energy separated from the other transitions, and can be easily obtained when, for instance, the sensitivity of the detector is increased. As an illustration, we have inserted the data obtained for 2.3 kbar in this figure.

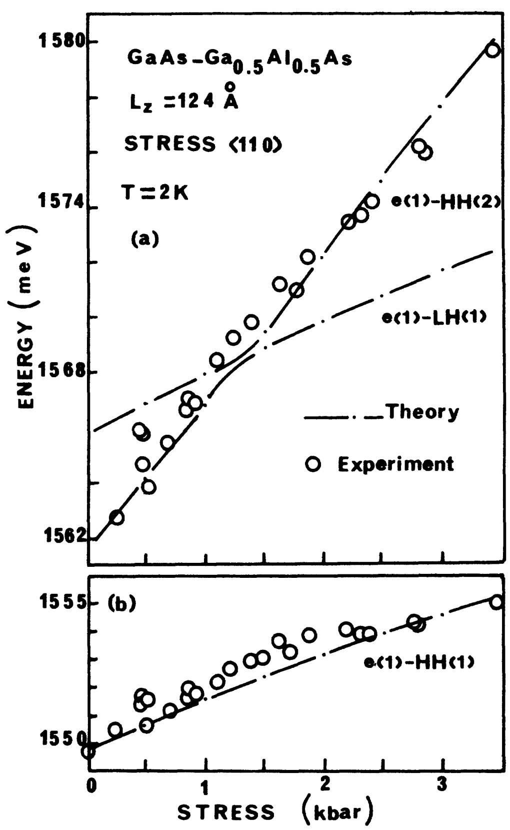

FIG. 4. Plot of the numerical calculation (dashed-dotted lines) obtained in the case of QW(1) ($L_z = 124$ Å) and as a function of the uniaxial stress. The transition energies have been calculated without taking the exciton binding energy into account. The experimental data (open circles) have been shifted from the Rydberg energy in order to correspond with the calculation. (a) Transition energy calculated between the first electron subband and the first light-hole one $e(1)$-LH(1) and the second heavy-hole subband $e(1)$-HH(2), and compared with the shift of the reflectance minima corresponding to LHE's. (b) The comparison between the shift of $e(1)$-HH(1) and the experimental data obtained for HHE's.

vestigation a large uncertainty is reducing the accuracy of the measurements in this part of the reflectance spectra. So, in the next section to check the agreement between the experiment and the theory, we will take the clearest data, those corresponding to the 31-Å-thick part of this series of reflectance patterns (2 and 2′ pair). To conclude this section, we will just point out the following. (i) In all cases, given a quantum well, LHE's and HHE's shift differently versus stress. (ii) The HHE's shift faster toward high energy than bulk GaAs, but the stress shift bends for a range of stress between 1 and 3.5 kbar (this is not the case for bulk GaAs for this range of stress.[1] The wider the well, the weaker the shift and the smaller the threshold for stress bending. (iii) The stress shift is larger for bulk GaAs than for the quantized-state LHE's; the wider the well, the stronger the shift.

IV. THEORETICAL RESULTS

We have calculated the effects of the in-plane [110] uniaxial stress for four different GaAs-$Al_{0.5}Ga_{0.5}As$ wells corresponding to 31, 53, 79, and 124 Å of well width. A six-band $\mathbf{k}\cdot\mathbf{p}$ model has been considered within the framework of the envelope-function approximation (EFA). The Pikus-Bir Hamiltonian[25] describes the effect of the uniaxial stress on the valence band. This model

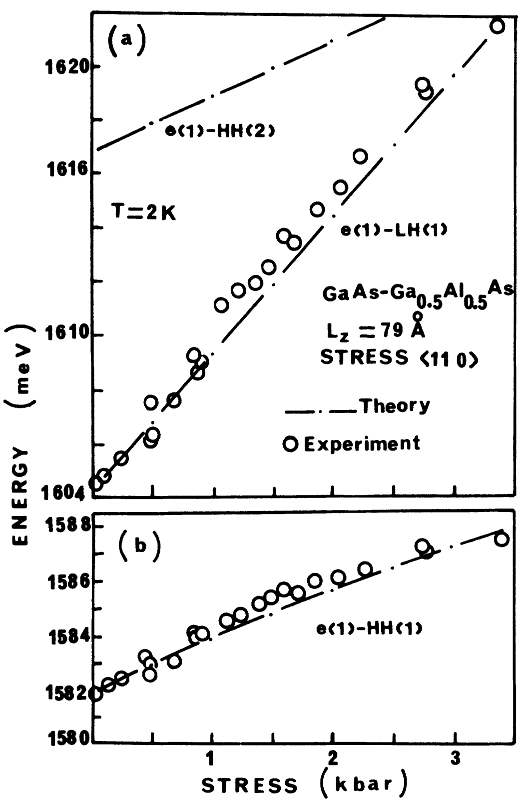

FIG. 5. Same as Fig. 4, but for $L_z = 79$ Å. In the range of stress concerned by this work the anticrossing between $e(1)$-HH(2) and $e(1)$-LH(1) was not obtained in contrast with the previous case.

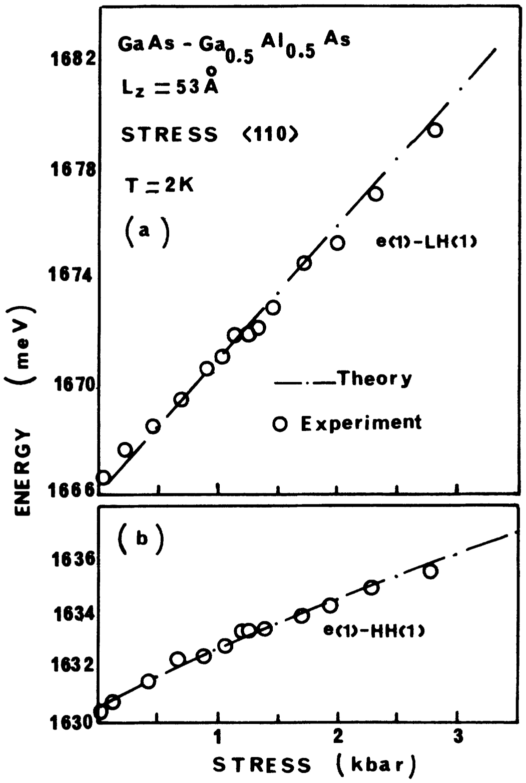

FIG. 6. Comparison between the band-to-band calculation and the experimental shifts when $L_z = 53$ Å. (a) Light-hole excitons. (b) Heavy-hole excitons.

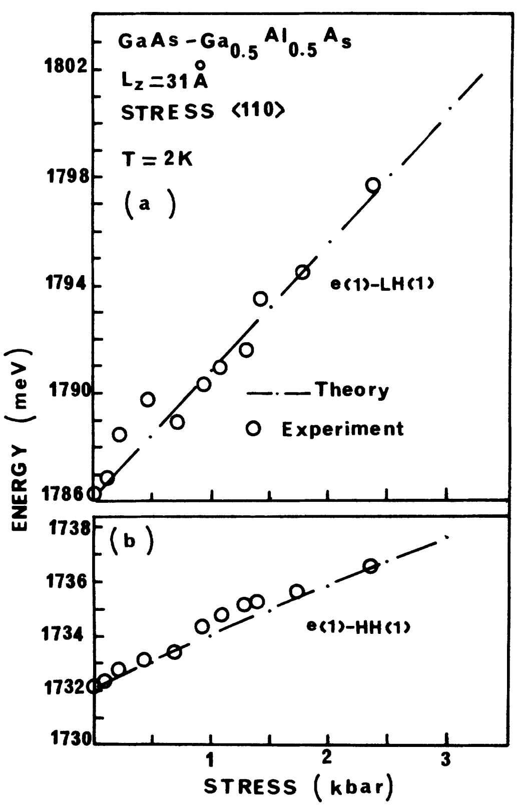

FIG. 7. The analog of Fig. 6 for $L_z = 31$ Å.

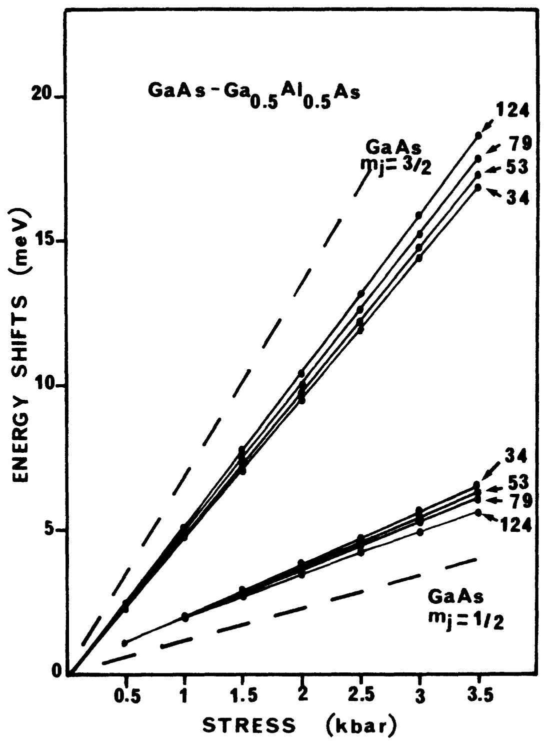

FIG. 8. Summary of the stress splitting of heavy-hole excitons (lower cluster) and light-hole excitons (upper cluster). For clarity in this figure, the experimental data have not been reported since they can be found in Figs. 4–7.

has already been proved[16,22,26] to give good results and is described in detail in Ref. 22.

The theoretical results are shown in Figs. 4–7 in comparison with the experiment. In Table I are given the Luttinger parameters (for the six-band model), compliance constants, conduction-band–valence-band coupling P parameter, and the deformation potentials considered in the calculation. Excitonic effects are not included in our model and the zero-stress energy of the transition is adjusted to the experimental result. Therefore it is only meaningful to compare the stress-induced shift of the transitions. The good agreement suggests that the assumption of stress-independent band offsets and excitonic binding energies is valid.[13,27] In spite of the good agreement between theory and experiment we observe a slightly higher bending in all the measured subbands as the stress increases, with respect to the calculated ones. It can be due to the fact that we do not consider the coupling with the split-off state, which can be more important for higher stress.[28,29]

TABLE I. Parameters used in calculation.

GaAs	$Al_{0.5}Ga_{0.5}As$
Luttinger parameters	
$\gamma_1 = 1.80$	1.40
$\gamma_2 = -0.42$	-0.48
$\gamma_3 = 0.38$	0.225
$P = 0.65$	0.65
Compliance constants (10^{-12} cm^2/dyn)	
$S_{11} = 1.16$	1.16
$S_{12} = -0.37$	-0.37
$S_{44} = 1.67$	1.67
Deformation potentials (eV)	
$a = 8.41$	8.41
$b = 1.76$	1.76
$d = 4.55$	4.55

Figure 8 summarizes the uniaxial stress dependencies of $e(1)$-HH(1) and $e(1)$-LH(1) transitions with respect to their zero-stress values, as a function of the well widths. Four quantum wells corresponding to 34, 53, 79, and 124 Å are analyzed. One can see, as expected, that when the well is thinner the mixed character of the subbands increases and the slopes of the two transitions, $e(1)$HH(1) and $e(1)$-LH(1), are closer to each other. Also, when the well is wider the slopes of these transitions are closer to their bulk values.

V. CONCLUSION

Reflectance spectroscopy performed at 2 K on four GaAs-$Ga_{0.5}Al_{0.5}As$ quantum wells, when a uniaxial stress is applied in the [110] direction up to 3.5 kbar, has allowed us to measure different stress dependencies for heavy-hole excitons and light-hole excitons. Moreover, the measured slopes have been found to be dependent on the well thickness. The experimental data have been explained by a theoretical calculation in the EFA.

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