

Strikingly Different Behaviors of Photoluminescence and Terahertz Generation in InGaN/GaN Quantum Wells

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Abstract—We have investigated photoluminescence (PL) and terahertz (THz) generation simultaneously from multiple InGaN/GaN quantum wells (QWs) with different well periods. The PL intensity fully saturates when the period of QWs is increased up to 4. However, THz output power continuously scales up even if the period of QWs is increased up to 16. Such a behavior indicates that high-power THz wave can be generated without efficient recombination of the photogenerated carriers, since THz is only generated during the absorption process. Following the measurements of intensity and peak energy of PL together with output power and spectra of THz, we have concluded that the screening effect induced by photo-generated carriers can be neglected when the pump fluence is as low as $85 \mu\text{J}/\text{cm}^2$.

Index Terms—Broadband terahertz (THz) wave, InGaN/GaN quantum wells (QWs), built-in field, dipole radiation, photoluminescence (PL).

I. INTRODUCTION

INGAN/GAN quantum wells (QWs) have attracted a lot of attention due to their potential applications in light-emitting devices covering the regions from blue to green [1]–[3]. Moreover, it was proposed that a quantum cascade laser based on nitrides could be operated at room temperature [4]. Perhaps, the most unique property of InGaN/GaN QWs is the presence of a strong built-in electric field in the order of several megavolts per centimeter originating from piezoelectric polarizations if the QWs are grown along the [0001] direction. Such a large built-in electric field induces a so-called quantum-confined Stark effect (QCSE), resulting in a significant separation of the electrons and holes generated in the InGaN/GaN QWs [5]–[7]. Due to such QCSE, the recombination rate is reduced since the poor overlap of wave functions of electron and hole. Obviously, for applications of laser diodes or light-emitting diodes, such electric field is an obstacle for achieving high efficiency or low

threshold devices. Therefore, in the past researchers have considered employing novel QWs structures in order to reduce the charge-separation effect [8], [9]. On the other hand, the built-in electric field in semiconductor bulk materials or nanostructures can be exploited for implementing certain devices without applying an external field. Indeed, terahertz (THz) radiation was generated from a GaAs surface being illuminated by ultrafast laser pulses due to the screening of surface depletion field by virtual photo-generated carriers [10]. In the past, THz emission from GaAs/AlGaAs QWs with an external electrical field being present was observed with its mechanism being attributed to creation of polarized electron–hole pairs by ultrafast laser pulses [11]. Recently, microwatt THz pulse was generated in an eight-period $\text{In}_{0.25}\text{Ga}_{0.75}\text{N}/\text{GaN}$ multiple QWs grown along [0001] direction by utilizing its internal field [12]. It was pointed out that for high pump fluence, screening effect induced by photo-generated carriers plays a significant role in THz generation process. Moreover, it is believed to be the main mechanism for saturation of THz output power generated in InGaN/GaN QWs [13], [14].

In this paper, we report our results obtained on InGaN/GaN QWs. Indeed, we simultaneously measured the photoluminescence (PL) and THz output in the InGaN/GaN QWs with different well periods. We have observed that the PL output was saturated when the period of QWs was increased up to four. However, the THz output scaled up more than linearly even with the period of the QWs increased up to 16. Through the comparison of experimental results of PL and THz measurements made on InGaN QWs with different well periods, we have concluded that high-power THz can be generated in InGaN QWs in spite of high density of nonradioactive structure defect. Such a conclusion is consistent with our proposed mechanism of THz generation in InGaN QWs, i.e., instantaneous generation of spatially separated electron–hole pairs resulting in efficient dipole radiation. Moreover, by combining the PL result with the THz signal, we have concluded that the screening effect induced by the photo-generated carriers is negligible for a pump fluence as low as $85 \mu\text{J}/\text{cm}^2$.

II. DESCRIPTION OF SAMPLES AND EXPERIMENTAL SETUP

InGaN/GaN QWs structures were grown on a $2.8\text{-}\mu\text{m}$ -thick unintentionally doped GaN template (the background electron density of $\sim 4 \times 10^{16} \text{ cm}^{-3}$) on a c-plane sapphire substrate by metal-organic chemical vapor deposition (MOCVD). The growth of the GaN template was performed at 1080°C by

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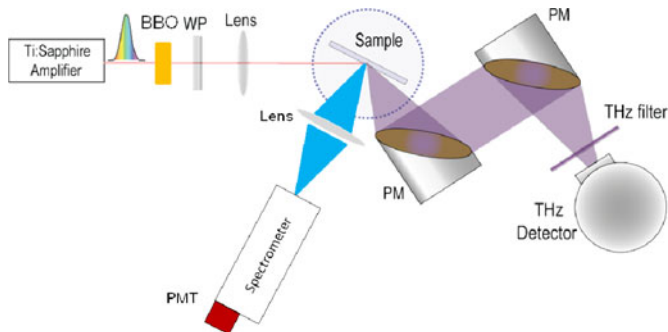


Fig. 1. Experimental setups used in the investigation of THz generation from InGaN/GaN quantum wells. WP: half-wave plate; PM: off-axis parabolic mirror; PMT: Photomultiplier. THz filters were used to block the residual pump laser beams while allowing the transmission of the THz waves.

employing 35-nm-thick low-temperature-grown ($T_g = 525^\circ\text{C}$) GaN buffer layer. Subsequently, $\text{In}_{0.19}\text{Ga}_{0.81}\text{N}/\text{GaN}$ QWs with different well periods ranging from 1 to 16 were deposited on the GaN template by MOCVD at 700°C . The thicknesses of the InGaN well and GaN barrier layers are 3 and 10 nm, respectively. Broadband THz pulses were generated by using a coherent radiation beam at the wavelength of 391 nm after frequency-doubling the output beam from a Ti:sapphire regenerative amplifier, see Fig. 1 for the experiment setups. The pulse duration is measured as 210 fs and repetition rate is 250 kHz. The excitation beam was then focused on the top surface of InGaN/GaN QWs with the laser spot area being measured to be about $0.85\text{ mm} \times 2.61\text{ mm}$. The polarization of laser was adjusted to be in the horizontal direction by using a half-wave plate, and incident angle was set as 70° to minimize the reflection of the pump beam. The THz radiation was collimated, and then, focused onto a 4.2-K Si bolometer or pyroelectric detector by a pair of gold-coated parabolic mirrors in reflection geometries. Simultaneously, the PL of the InGaN QWs, excited by the same pump beam, was collected by a lens and then measured by a photomultiplier (PMT) after passing through a single-channel high-resolution spectrometer.

III. RESULTS AND DISCUSSIONS

In this section, we present our results obtained on the THz generation and PL from the $\text{In}_{0.19}\text{Ga}_{0.81}\text{N}/\text{GaN}$ QWs with different periods.

We have first investigated the PL spectra of each sample under room temperature with the pump fluence being set to $85\ \mu\text{J}/\text{cm}^2$. As shown in Fig. 2(a), the emission spectrum measured on each sample peaks around 472 nm. The multiple peaks modulation is caused by Fabry-Pérot resonance due to the multiple reflection caused by the front and back surfaces of each sample. From the spectra, it is clear that PL intensity scales up when the well period is increased up to four. However, when we further increase the well period, the PL intensity is significantly saturated, see Fig. 2(b). According to [15], we estimated the absorption length of the InGaN QW layers in our QW structure to be on the order of 100 nm. Since the total thickness of the InGaN layers in the 16 periods of the InGaN/GaN QWs is 48 nm, we have ruled out

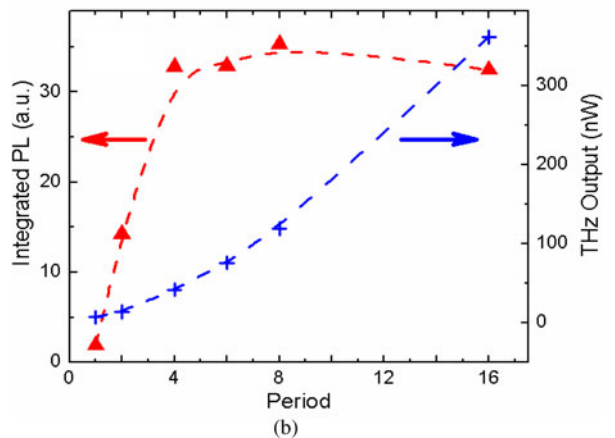
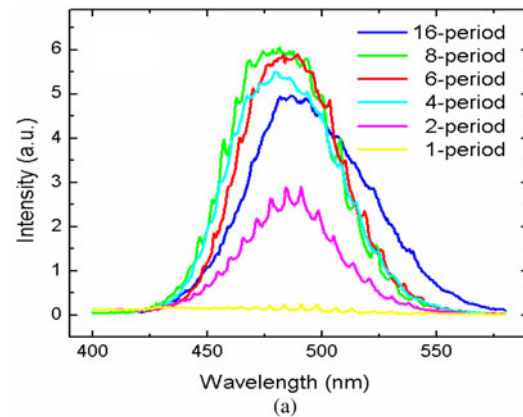


Fig. 2. (a) PL spectra of InGaN quantum wells with different periods measured at room temperature. (b) PL intensity and THz output power versus well period. Red triangular corresponds to PL intensity and blue cross corresponds to THz output power. The pump fluence is set as $85\ \mu\text{J}/\text{cm}^2$.

the penetration saturation of the optical pumping as the mechanism for the PL saturation. In fact, for the 16-period sample, the transmitted pump beam is visible to the naked eyes. Such saturation could be caused by the increased density of the non-radiative structure defects. When the total thickness of the QWs scales up, the accumulated strain is expected to increase, which results in a higher density of nonradiative structure defects. It is worth noting that for the 16-period InGaN QWs, we have clearly observed submillimeter dots by naked eyes within the laser beam spot, especially on the edge of sample. These dots can be the indication that the total thickness of the InGaN/GaN QWs in this sample is close to the critical thickness.

For the THz generation, as illustrated by Fig. 2(b), the output power increases more than linearly even if the period of the QWs is increased up to 16. The highest output power collected from the 16-period InGaN QWs is 360 nW. It is worth noting that the THz output power is not reduced when we move the laser spot onto the location where the sub-millimeter dots are observed. In our recent paper [12], we have attributed the mechanism for the THz generation to instantaneous creation of spatially separated electron-hole pairs resulting in efficient dipole radiation. It is well known that the output power for dipole radiation is proportional to square of dipole density. The density of the photogenerated carriers is proportional to the intensity of the

optical pump. In addition, there are no phase differences among the THz waves generated by different QWs along the propagation direction. If we assume that the absorption is constant in each well, the THz output power should be quadratically dependent on the number of the QWs. The assumption that the phase difference of the generated THz wave in each QW is negligible makes sense since the total thickness of QWs is much smaller than the wavelength of the THz wave. Furthermore, unlike PL, the THz wave is generated during the absorption process not during the radiation process in the InGaN QWs. Since the non-radiative structure defects only influence the radiation process, the THz output power continuously increases with increasing the period of the QWs even if the PL exhibits saturation. Thus, in principle, we can further increase the periods of the InGaN QWs to further scale up the THz output power. The increase in the density of the nonradiative defects only influences the recombination process. For example, if we can add another 16 periods, the THz output power can be enhanced by a factor of 4. The increase in the indium composition in the wells can also be used to increase the THz output power since it causes electrons holes to further separate spatially, and therefore, the dipole strength can be further increased. Under the same experimental setup, an eight-period $\text{In}_{0.25}\text{Ga}_{0.75}\text{N}/\text{GaN}$ QWs was measured, which shows less than a half of the PL intensity but nine times more THz output power than that of the eight-period $\text{In}_{0.19}\text{Ga}_{0.81}\text{N}/\text{GaN}$ QWs described in this paper. Previously, it has been shown that by increasing the well width from 1.8 to 3.6 nm, THz output power can be increased by 30% [13]. By optimizing the structure of the QWs, the THz output power is expected to increase to as high as $20 \mu\text{W}$ under our experiment condition.

The dependence of the THz output power on the pump fluence for the 16-period InGaN QWs is shown in Fig. 3(a). Initially, for the pump fluencies up to $40 \mu\text{J}/\text{cm}^2$, the THz output power quadratically increases with the pump fluence, see the red fitting curve in Fig. 3(a). When the pump fluence is further increased up to $85 \mu\text{J}/\text{cm}^2$, a slight deviation to the quadratic fit is observed. Such a deviation has been attributed to the screening effect induced by the photo-generated electron-hole pairs and been supported by blueshift of the PL peak energy [13]. We have also plotted the peak energy of PL versus the pump fluence, see the blue dots of Fig 3(b). The peak energy for such 16-period InGaN QWs exhibits a blueshift in the amount of 60 meV, which appears to support the screening effect. However, according to the theoretical calculation [14], the screening effect increases the effective absorption coefficient, which is proportional to ratio of the integrated PL intensity with the pump fluence. Thus, based on such a calculation, we should expect that the integrated PL intensity scales up more than linearly when the pump fluence is increased. In experiment, apparently, the integrated PL intensity scales up less than linearly with the pump fluence, see the red triangles of Fig. 3(a), indicating that the absorption coefficient is actually reduced, which is not consistent with the screening effect. In a recent pump-probe experiment on similar InGaN QWs, the reduction of absorption has been observed for a pump fluence of as low as $0.19 \text{ mJ}/\text{cm}^2$ and explained by increased density of the hot carriers [16]. Thus, from the dependence of

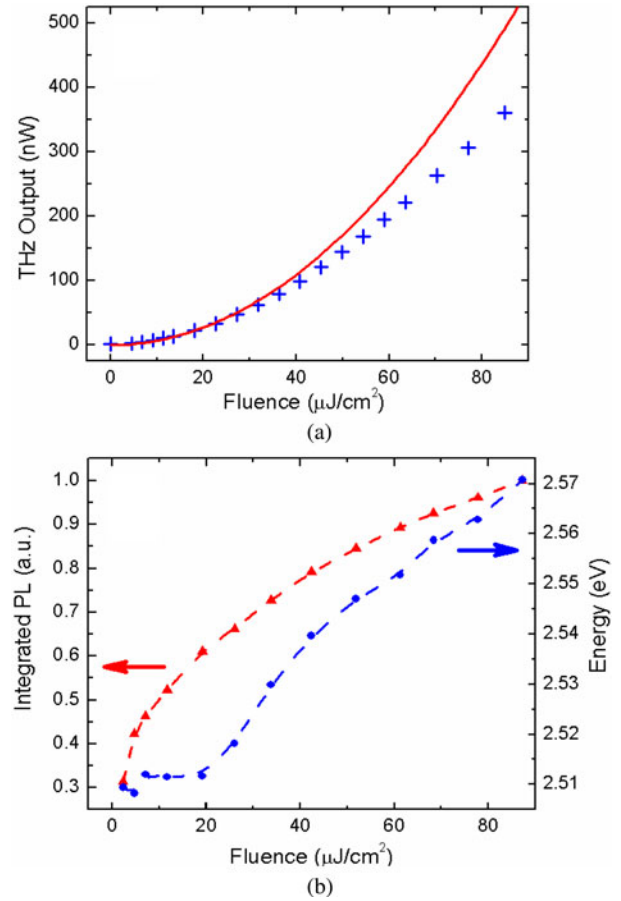


Fig. 3. (a) THz output power as a function of pump fluence. The crosses correspond to experiment data. The red curve corresponds to a quadratic fitting for first ten data points. (b) Integrated PL intensity and peak energy as a function of pump fluence. The red triangles and blue dots correspond to PL intensity and peak energy, respectively.

the integrated PL intensity on the pump fluence, we may draw a conclusion that the deviation or saturation is induced by the decrease in the absorption coefficient.

The contradicting conclusions from our analysis of the PL reveal that monitoring the PL signal may not be sufficient for us to understand the dynamic screening effect in the THz generation process, since THz is only generated during the absorption process whereas the PL is not only influenced by the absorption process but also by the recombination process. Indeed, the recombination mechanism is still not fully understood in the InGaN/GaN QWs. It is widely believed that the localized states play an important role in the emission process [17]–[19]. Such localized states induce a band-tail filling effect, resulting in the blue shift with increasing the pump power. Therefore, the blueshift for the InGaN QWs has been attributed to the combination of screening effect with band-tail filling effect [20]. In our detailed PL study on a similar InGaN sample, the dominant PL peak is explained by the recombination of the localized states [21]. Thus, we may over-emphasize the role of the screening effect if only monitoring the blueshift of PL peak.

An alternative method to determine the screening effect is to measure the spectra of the THz output from the InGaN QWs at

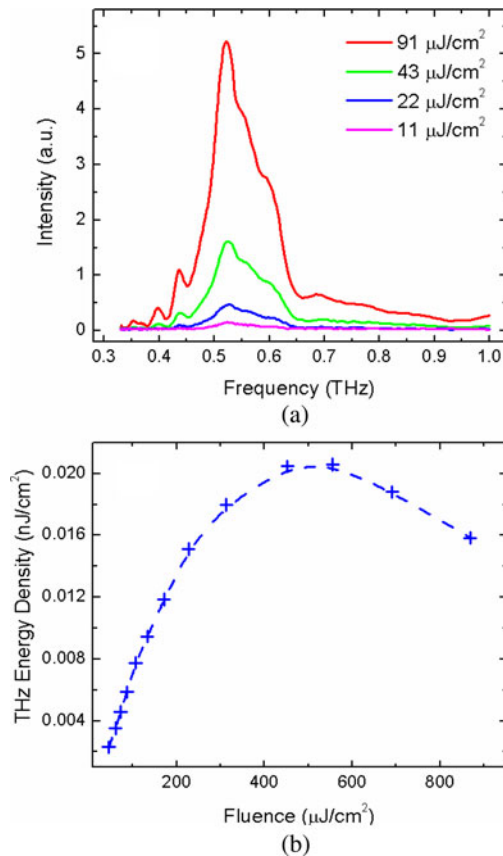


Fig. 4. (a) THz spectra measured at pump fluences 11, 22, 43, 91 $\mu\text{J}/\text{cm}^2$. (b) THz energy density generated per well as function of pump fluence.

different pump fluencies. It has been pointed out in theory that dramatic broadening and shift of the THz spectra will be observed with increasing the pump fluence if dynamical screening is a strong absorption process [14]. Under such a predication, the THz spectral band width could far more exceed the bandwidth of the excitation laser. We have measured the THz spectra under several different pump fluencies by a homemade sub-millimeter diffraction grating system. Such a system allows us to detect whole THz spectra even if the bandwidth of the THz output is beyond the laser spectra, which is difficult to achieve in an electro-optic sampling system if the same laser is used to generate and sample the THz output. As is shown in Fig. 4(a), when the pump fluence is increased from 10 to 90 $\mu\text{J}/\text{cm}^2$, within the accuracy of our measurements, we did not observe a bandwidth broadening or frequency shift of the THz spectra, which proves that in our experiment the screening effect induced by the photogenerated electron-hole pairs is negligible. It is worth noting that our conclusion is not in conflict with the work in [14], since the lowest fluence used in their calculation, which only causes a weak screening effect, is about two times as large as the highest value in our experiment.

In order to see a strong screening effect, we have reduced laser spot area to increase the pump fluence up to 1 mJ/cm^2 . A full saturation behavior of the THz output power was observed when the pump fluence is about 0.6 mJ/cm^2 . This saturation

pump value is agreed with [16]. Because of the screening effect, the highest THz energy that can be generated in a single pulse is limited by the total static energy originally stored in the InGaN/GaN QWs. According to the simple capacitor model [13], the total energy stored in our InGaN QWs is estimated to be at least 100 nJ/cm^2 . Experimentally, the saturation value for the THz energy density generated on each well is only about 0.02 nJ/cm^2 , see Fig. 4(b), which is a factor of 5000 lower than the theoretical value. Thus, the screening effect is not the only mechanism to limit the THz output. A large amount THz output may not be collected in our experimental setup since a large fraction of the THz beam undergoes total internal reflection at the sample surface. Indeed, since the built-in field is always perpendicular to the sample surface, the most efficient radiation direction is along the surface, which can be hardly coupled out of the sample. In addition, we believe that free carriers inside the GaN template or photogenerated carriers inside the QWs also absorb a significant amount of the THz output power. Our simplified calculation indicates that the collected output power of 360 nW obtained on the 16-period QWs corresponds to the total output power of about 60 μW generated by the QWs. Previously, the increase in the absorption of the THz wave by the photogenerated carriers was investigated and evidenced in GaTe and InN materials [22], [23]. Such a mechanism can also explain the reduction of the THz output power when the pump fluence is higher than 0.6 mJ/cm^2 , as shown in Fig. 4(b). Under the condition that the maximum degree of the screening effect is achieved, if increasing the pump fluence, more carriers are generated but there is no further increase in the THz output power. Since the more carriers only increase the absorption, the THz output power is reduced.

IV. CONCLUSION

In conclusion, we have simultaneously investigated the PL and THz output from a set of the InGaN/GaN QWs with different periods. Since it is only generated during the absorption process, the THz output power continuously scales up with the number of the QWs. Thus, we can further increase the THz output power by adding up more periods of the QWs or increasing the indium composition in the well region. In contrast, the photoluminescence intensity may not increase. By measuring the THz spectra, we have concluded that for the pump fluence as low as 85 $\mu\text{J}/\text{cm}^2$, the screening effect induced by the photo-generated carriers is negligible.

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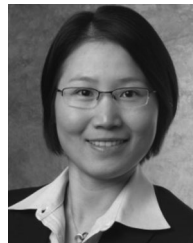


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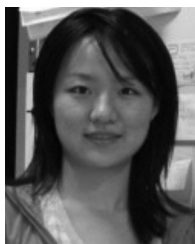
Dr. Zhao received the SPIE Educational Scholarship in optical science and engineering for the years of 2008 and 2009. She also served as a panel review member for the National Science Foundation.



Guangyu Liu received the B.S. degree in electronic science and technology from the Huazhong University of Science and Technology, Hubei, China, in June 2008. Since July 2008, she has been working toward the Ph.D. degree in the Department of Electrical and Computer Engineering (ECE) and the Center for Optical Technologies (COT), Lehigh University, Bethlehem, PA.

Her research interests are related to III-Nitride semiconductor nanostructures and optoelectronics devices, covering the theoretical/computational, metalorganic chemical vapor deposition (MOCVD) growth, and device fabrication and characterization for high-performance III-Nitride light-emitting diodes (LEDs), III-Nitride quantum dots.

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Dr. Tansu was the Wisconsin Alumni Research Foundation Graduate University Fellow and the Vilas Graduate University Fellow during his graduate studies at Wisconsin. He received the Graduate Dissertator Award at Wisconsin. Other selected awards include: the Harold A. Peterson Best ECE Dissertation Award (at Wisconsin), the 2008 Libsch Early Career Research Award (at Lehigh), and the 2010 Wisconsin Forward Under 40 for the Outstanding Young Alumni Award (at Wisconsin). He has also reviewed regularly with the leading journals in applied physics, quantum electronics, nanotechnology, photonics, and optoelectronics areas. He has also served several times as a panel member for the U.S. National Science Foundation (ECCS, DMR, STC Panel, ERC Panel/Site Reviewer, and SBIR), U.S. Department of Defense, U.S. Department of Energy, several other U.S. federal agencies, private foundations in U.S., and several funding agencies in Europe and Asia. Previously, he has also given numerous lectures, seminars, and invited talks (total >40) in universities, research institutions, and conferences in U.S., Canada, Europe, and Asia. He serves as the Primary Guest Editors of the IEEE JOURNAL OF SELECTED TOPICS IN QUANTUM ELECTRONICS (2008–2009) and the IEEE/OSA JOURNAL OF DISPLAY TECHNOLOGY (2012–2013), and he also serves as Associate Editors for IEEE PHOTONICS JOURNAL (2009–present), *OSA Optical Materials Express* (2011–present), and *Nanoscale Research Letters* (2007–present). He has also served as the Technical Program Committee for several major technical conferences for IEEE, OSA, SPIE, and APS; the selected lists include: IEEE/OSA Conference on Lasers and Electro-Optics (2007, 2008, 2009, and 2013), SPIE PhotonicsWest (2009–2013), APS March Annual Meeting (2007, 2009, and 2010), and others. He was also selected as Invited General Participants at the 2008 National Academy of Engineering (NAE)'s U.S. Frontiers of Engineering (USFOE) Symposium and the NAE's 2012 German-American Frontiers of Engineering Symposium (GAFOE), and he served as the Organizing Committee for the 2009 NAE's U.S. Frontiers of Engineering Symposium.