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# analytical chemistry

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**Magnetically-Assisted Surface  
Enhanced Raman Spectroscopy for  
Label-Free Determination of Human  
Immunoglobulin G in Blood Using  
 $\text{Fe}_3\text{O}_4$ @Ag Nanocomposite**



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**ON THE COVER:** Schematic representation of magnetically assisted surface enhanced Raman spectroscopy (MA-SERS) determination of immunoglobulins (IgG) using magnetite nanoparticles (brown) with attached silver nanoclusters (yellow). The helix represents streptavidin molecule acting as a linker for antigen immobilization. Image created by Iva Hrubošová.

## Perspectives

10979 

DOI: 10.1021/ac5027435

**A Mass-Spectrometry-Based Framework To Define the Extent of Disorder in Proteins**

Rebecca Beveridge, Sam Covill, Kamila J. Pacholarz, Jason M. D. Kalapothakis, Cait E. MacPhee, and Perdita E. Barran\*

## Letters to Analytical Chemistry

10992 

DOI: 10.1021/ac5025495

**Homogeneous Electrochemical Assay for Protein Kinase Activity**

Ik-Soo Shin, Rohit Chand, Sang Wook Lee, Hyun-Woo Rhee, Yong-Sang Kim,\* and Jong-In Hong\*

10996 

DOI: 10.1021/ac502779r

**Approaching Sensitivity of Tens of Ions Using Atomically Precise Cluster–Nanofiber Composites**

Atanu Ghosh, Vedhakkani Jeseentharani, Mohd Azhardin Ganayee, Rani Gopalakrishnan Hemalatha, Kamallesh Chaudhari, Cherianath Vijayan, and Thalappil Pradeep\*

11002 

DOI: 10.1021/ac503280v

**Strategy for Characterization of Distribution and Associations of Organobromine Compounds in Soil Using Synchrotron Radiation Based Spectromicroscopies**

Lei Luo, Jitao Lv, Chuang Xu, and Shuzhen Zhang\*

11006 

DOI: 10.1021/ac503439n

**Sheathless Capillary Electrophoresis-Tandem Mass Spectrometry for Top-Down Characterization of *Pyrococcus furiosus* Proteins on a Proteome Scale**

Xuemei Han, Yueju Wang, Aaron Aslanian, Marshall Bern, Mathieu Lavallée-Adam, and John R. Yates III\*

11013 

DOI: 10.1021/ac503453v

**Single-Cell Copy Number Analysis of Prostate Cancer Cells Captured with Geometrically Enhanced Differential Immunocapture Microdevices**

Erica D. Pratt, Asya Stepansky, James Hicks, and Brian J. Kirby\*

## Technical Notes

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11018 

DOI: 10.1021/ac502365w

### High Precision Fractionator for Use with Density Gradient Ultracentrifugation

Yara Kadria-Vili, Griffin Canning, Sergei M. Bachilo, and R. Bruce Weisman\*

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11024

DOI: 10.1021/ac502511a

### LipidBlast Templates As Flexible Tools for Creating New in-Silico Tandem Mass Spectral Libraries

Tobias Kind,\* Yozo Okazaki, Kazuki Saito, and Oliver Fiehn\*

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11028

DOI: 10.1021/ac502671f

### Enhanced Detection of DNA Sequences Using End-Point PCR Amplification and Online Gel Electrophoresis (GE)-ICP-MS: Determination of Gene Copy Number Variations

T. Iglesias González, M. Espina, L. M. Sierra, J. Bettmer, E. Blanco-González,\* M. Montes-Bayón,\* and A. Sanz-Medel

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11033 

DOI: 10.1021/ac502883p

### Assessment of Serum Protein Dynamics by Native SILAC Flooding (SILflood)

Hendrik Nolte, Soraya Hölper, Matthias Selbach, Thomas Braun, and Marcus Krüger\*

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11038 

DOI: 10.1021/ac5029209

### All-Solid-State Polymeric Membrane Ion-Selective Miniaturized Electrodes Based on a Nanoporous Gold Film as Solid Contact

Tanji Yin, Dawei Pan, and Wei Qin\*

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## Articles

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11045 

DOI: 10.1021/ac500094v

### Determination of Multivalent Protein–Ligand Binding Kinetics by Second-Harmonic Correlation Spectroscopy

Krystal L. Sly and John C. Conboy\*

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11055 

DOI: 10.1021/ac500767m

### Improved Methylene Blue Two-Phase Titration Method for Determining Cationic Surfactant Concentration in High-Salinity Brine

Leyu Cui, Maura Puerto, José L. López-Salinas, Sibani L. Biswal,\* and George J. Hirasaki\*

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11062 

DOI: 10.1021/ac5031804


### Compact Hybrid (Gold Nanodendrite-Quantum Dots) Assembly: Plasmon Enhanced Fluorescence-Based Platform for Small Molecule Sensing in Solution

Huide Chen and Yunsheng Xia\*

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- 11070  DOI: 10.1021/ac503323e  
**Automated Dispersive Solid-Phase Extraction Using Dissolvable Fe<sub>3</sub>O<sub>4</sub>-Layered Double Hydroxide Core–Shell Microspheres as Sorbent**  
Sheng Tang, Guo Hui Chia, Yuepeng Chang, and Hian Kee Lee\*
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- 11077  DOI: 10.1021/ac503425g  
**Enhanced Single Molecule Mass Spectrometry via Charged Metallic Clusters**  
Christopher E. Angevine, Amy E. Chavis, Nuwan Kothalawala, Amala Dass, and Joseph E. Reiner\*
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- 11086  DOI: 10.1021/ac503171f  
**Analysis of *Drosophila* Lipids by Matrix-Assisted Laser Desorption/Ionization Mass Spectrometric Imaging**  
Ann-Christin Niehoff, Hans Kettling, Alexander Pirkl, Yin Ning Chiang, Klaus Dreisewerd, and Joanne Y. Yew\*
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- 11093  DOI: 10.1021/ac5025478  
**Label-Free Biochemical Analytic Method for the Early Detection of Adenoviral Conjunctivitis Using Human Tear Biofluids**  
Samjin Choi,\* Sung Woon Moon, Jae-Ho Shin, Hun-Kuk Park, and Kyung-Hyun Jin\*
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- 11100  DOI: 10.1021/ac503360n  
**A Label-Free Aptamer-Fluorophore Assembly for Rapid and Specific Detection of Cocaine in Biofluids**  
Daniel Roncancio, Haixiang Yu, Xiaowen Xu, Shuo Wu, Ran Liu, Joshua Debord, Xinhui Lou, and Yi Xiao\*
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- 11107  DOI: 10.1021/ac503347h  
**Magnetically-Assisted Surface Enhanced Raman Spectroscopy (MA-SERS) for Label-Free Determination of Human Immunoglobulin G (IgG) in Blood Using Fe<sub>3</sub>O<sub>4</sub>@Ag Nanocomposite**  
Anna Balzerova, Ariana Fargasova, Zdenka Markova, Vaclav Ranc,\* and Radek Zboril\*
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- 11115  DOI: 10.1021/ac502053s  
**In Situ Determination of Colloidal Gold Concentrations with UV–Vis Spectroscopy: Limitations and Perspectives**  
Thomas Hendel, Maria Wuithschick, Frieder Kettemann, Alexander Birnbaum, Klaus Rademann,\* and Jörg Polte\*
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- 11125  DOI: 10.1021/ac502082b  
**Detecting Exosomes Specifically: A Multiplexed Device Based on Alternating Current Electrohydrodynamic Induced Nanoshearing**  
Ramanathan Vaidyanathan, Maedeh Naghibosadat, Sakandar Rauf, Darren Korbie, Laura G. Carrascosa, Muhammad J. A. Shiddiky,\* and Matt Trau\*
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- 11133  DOI: 10.1021/ac5038329  
**Monitoring Antibody Aggregation in Early Drug Development Using Raman Spectroscopy and Perturbation–Correlation Moving Windows**  
Ramón Gómez de la Cuesta, Royston Goodacre, and Lorna Ashton\*

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11141  DOI: 10.1021/ac502197u  
**High-Precision  $^{143}\text{Nd}/^{144}\text{Nd}$  Ratios from  $\text{NdO}^+$  Data Corrected with in-Run Measured Oxygen Isotope Ratios**  
Zhu-Yin Chu,\* Chao-Feng Li, Ernst Hegner, Zhi Chen, Yan Yan, and Jing-Hui Guo

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11151  DOI: 10.1021/ac502138p  
**Transmission Geometry Laser Desorption Atmospheric Pressure Photochemical Ionization Mass Spectrometry for Analysis of Complex Organic Mixtures**  
Leonard Nyadong, Mmili M. Mapolelo, Christopher L. Hendrickson, Ryan P. Rodgers,\* and Alan G. Marshall\*

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11159  DOI: 10.1021/ac5034177  
**Coupling Solid-Phase Microextraction with Ambient Mass Spectrometry Using Surface Coated Wooden-Tip Probe for Rapid Analysis of Ultra Trace Perfluorinated Compounds in Complex Samples**  
Jiewei Deng, Yunyun Yang, Ling Fang, Li Lin, Haiyun Zhou, and Tiangang Luan\*

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11167 DOI: 10.1021/ac5024292  
**Development of Particle Induced Gamma-Ray Emission Methods for Nondestructive Determination of Isotopic Composition of Boron and Its Total Concentration in Natural and Enriched Samples**  
Sumit Chhillar, Raghunath Acharya,\* Suparna Sodaye, and Pradeep K. Pujari

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11174 DOI: 10.1021/ac502596m  
**Electroosmotic Flow in Nanofluidic Channels**  
Daniel G. Haywood, Zachary D. Harms, and Stephen C. Jacobson\*

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11181  DOI: 10.1021/ac502600a  
**Quantum Dot-Based Concentric FRET Configuration for the Parallel Detection of Protease Activity and Concentration**  
Miao Wu, Eleonora Petryayeva, and W. Russ Algar\*

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11189 DOI: 10.1021/ac502631z  
**Characterization of an Ion Sensitive Field Effect Transistor and Chloride Ion Selective Electrodes for pH Measurements in Seawater**  
Yuichiro Takeshita, Todd R. Martz,\* Kenneth S. Johnson, and Andrew G. Dickson

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11196  DOI: 10.1021/ac5026489  
**Real-Time Fluorescence Tracking of Gene Delivery via Multifunctional Nanocomposites**  
Min Bai, Xilin Bai, and Leyu Wang\*

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11203  DOI: 10.1021/ac502670t  
**Combined Raman Microspectrometer and Shearforce Regulated SECM for Corrosion and Self-Healing Analysis**  
Mathieu Etienne,\* Manuel Dossot, Jérôme Grausem, and Grégoire Herzog

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11211 DOI: 10.1021/ac502705n

**Highly Sensitive Detection of DNA Hybridization on Commercialized Graphene-Coated Surface Plasmon Resonance Interfaces**

Oleksandr Zagorodko, Jolanda Spadavecchia, Aritz Yanguas Serrano, Iban Larroulet, Amaia Pesquera, Amaia Zurutuza, Rabah Boukherroub, and Sabine Szunerits\*

11217 DOI: 10.1021/ac502774m

**Experimental and Numerical Study of Submonolayer Sputter Deposition of Polystyrene Fragments on Silver for the Storing Matter Technique**

Canan Turgut, Godhuli Sinha, Lotta Mether, Jouko Lahtinen, Kai Nordlund, Mohammed Belmahi, and Patrick Philipp\*

11226 DOI: 10.1021/ac502776z

**Molecular Scavengers as Carriers of Analytes for Mass Spectrometry Identification**

Marek Smoluch,\* Michal Ceglowski, Joanna Kurczewska, Michal Babij, Teodor Gotszalk, Jerzy Silberring, and Grzegorz Schroeder

11230 DOI: 10.1021/ac5027836

**Ion-Transfer Voltammetry of Perfluoroalkanesulfonates and Perfluoroalkancarboxylates: Picomolar Detection Limit and High Lipophilicity**

Mohammed B. Garada, Benjamin Kabagambe, Yushin Kim, and Shigeru Amemiya\*

11238 DOI: 10.1021/ac5028512

**Improving the Quantification of Secondary Organic Aerosol Using a Microflow Reactor Coupled to HPLC-MS and NMR to Manufacture Ad Hoc Calibration Standards**

Emanuela Finessi, Richard T. Lidster, Fiona Whiting, Thomas Elliott, M. Rami Alfarra, Gordon B. McFiggans, and Jacqueline F. Hamilton\*

11246 DOI: 10.1021/ac503142u

**The Use of Ion Mobility Mass Spectrometry for Isomer Composition Determination Extracted from Se-Rich Yeast**

Johann Far, Cédric Delvaux, Christopher Kune, Gauthier Eppe, and Edwin de Pauw\*

11255 DOI: 10.1021/ac5038957

**Complementary Analytical Liquid Chromatography Methods for the Characterization of Aqueous Phase from Pyrolysis of Lignocellulosic Biomasses**

Débora Tomasini, Francesco Cacciola, Francesca Rigano, Danilo Sciarrone, Paola Donato, Marco Beccaria, Elina B. Caramão, Paola Dugo, and Luigi Mondello\*

11263 DOI: 10.1021/ac502843t

**Visual and Portable Strategy for Copper(II) Detection Based on a Striplike Poly(Thymine)-Caged and Microwell-Printed Hydrogel**

Zhihe Qing, Zhengui Mao, Taiping Qing, Xiaoxiao He,\* Zhen Zou, Dinggeng He, Hui Shi, Jin Huang, Jianbo Liu, and Kemin Wang\*

11269 

DOI: 10.1021/ac502845b

**Sensitive Detection of DNA Methyltransferase Activity Based on Exonuclease-Mediated Target Recycling**

Xi-Wen Xing, Feng Tang, Jun Wu, Jie-Mei Chu, Yu-Qi Feng, Xiang Zhou,\* and Bi-Feng Yuan\*

11275

DOI: 10.1021/ac502897g

**Depth Profiling of Element Concentrations in Stratified Materials by Confocal Microbeam X-ray Fluorescence Spectrometry with Polychromatic Excitation**

Pawel Wrobel,\* Dariusz Wegrzynek, Mateusz Czyzycki, and Marek Lankosz

11281 

DOI: 10.1021/ac502943d

**Identification of the Reactive Metabolites of Fenclozic Acid in Bile Duct Cannulated Rats**

Scott Martin,\* Eva M. Lenz, Warren Keene, and Malcolm R. Clench

11290 

DOI: 10.1021/ac5030717

***In Situ* Electrochemical Stress Measurements Examining the Oxygen Evolution Reaction in Basic Electrolytes**

Thao T. H. Hoang, Yair Cohen, and Andrew A. Gewirth\*

11298 

DOI: 10.1021/ac503000e

**ChelomEx: Isotope-Assisted Discovery of Metal Chelates in Complex Media Using High-Resolution LC-MS**

Oliver Baars,\* François M. M. Morel, and David H. Perlman

11306 

DOI: 10.1021/ac503026d

**Gadolinium Oxide Nanoparticles and Aptamer-Functionalized Silver Nanoclusters-Based Multimodal Molecular Imaging Nanoprobe for Optical/Magnetic Resonance Cancer Cell Imaging**

Jingjing Li, Jia You, Yue Dai, Meilin Shi, Cuiping Han, and Kai Xu\*

11312

DOI: 10.1021/ac5030478

**Tunable near-Infrared Luminescence of PbSe Quantum Dots for Multigas Analysis**

Long Yan, Yu Zhang,\* Tieqiang Zhang, Yi Feng, Kunbo Zhu, Dan Wang, Tian Cui, Jingzhi Yin, Yiding Wang, Jun Zhao, and William W. Yu\*

11319

DOI: 10.1021/ac5030528

**Design and Optimization of a Total Vaporization Technique Coupled to Solid-Phase Microextraction**

Christina L. Rainey, Dana E. Bors, and John V. Goodpaster\*

11326 

DOI: 10.1021/ac5030667

**Multiplexed Analysis of Genes and of Metal Ions Using Enzyme/DNAzyme Amplification Machineries**

Lina Freage, Fuan Wang, Ron Orbach, and Itamar Willner\*

- 11334  DOI: 10.1021/ac503067a  
**Kinetics of Precursor Labeling in Stable Isotope Labeling in Cell Cultures (SILAC) Experiments**  
Tian Zhang, John C. Price, Eslam Nouri-Nigjeh, Jun Li, Marc K. Hellerstein, Jun Qu, and Sina Ghaemmaghami\*
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- 11342  DOI: 10.1021/ac5030842  
**Enhanced Electroanalysis in Lithium Potassium Eutectic (LKE) Using Microfabricated Square Microelectrodes**  
Damion K. Corrigan, Ewen O. Blair, Jonathan G. Terry, Anthony J. Walton, and Andrew R. Mount\*
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- 11349  DOI: 10.1021/ac5035049  
**All-in-One Centrifugal Microfluidic Device for Size-Selective Circulating Tumor Cell Isolation with High Purity**  
Ada Lee, Juhee Park, Minji Lim, Vijaya Sunkara, Shine Young Kim, Gwang Ha Kim, Mi-Hyun Kim, and Yoon-Kyoung Cho\*
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- 11357  DOI: 10.1021/ac503255f  
**Selective and Sensitive Turn-on Chemosensor for Arsenite Ion at the ppb Level in Aqueous Media Applicable in Cell Staining**  
Somenath Lohar, Siddhartha Pal, Buddhadeb Sen, Manjira Mukherjee, Samya Banerjee, and Pabitra Chattopadhyay\*
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- 11362  DOI: 10.1021/ac503300y  
**Barcode-Like Paper Sensor for Smartphone Diagnostics: An Application of Blood Typing**  
Liyun Guan, Junfei Tian, Rong Cao, Miaosi Li, Zhaoxiang Cai, and Wei Shen\*
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- 11368  DOI: 10.1021/ac503318e  
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T. S. Biswas, N. Miriyala, C. Doolin, X. Liu, T. Thundat, and J. P. Davis\*
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- 11373  DOI: 10.1021/ac5033319  
**Electrogenerated Chemiluminescence Resonance Energy Transfer between Luminol and CdSe@ZnS Quantum Dots and Its Sensing Application in the Determination of Thrombin**  
Yong-Ping Dong, Ting-Ting Gao, Ying Zhou, and Jun-Jie Zhu\*
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- 11380  DOI: 10.1021/ac503334a  
**Comparison of Sample Introduction Methods for Continuous Chemical Purification in Two-Dimensional Electro-Fluid-Dynamic Devices**  
Chang Liu and David D. Y. Chen\*
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- 11387  DOI: 10.1021/ac503344f  
**Exhaustive Thin-Layer Cyclic Voltammetry for Absolute Multianalyte Halide Detection**  
María Cuartero, Gastón A. Crespo, Majid Ghahraman Afshar, and Eric Bakker\*



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11396  DOI: 10.1021/ac503379e

**A Study of Calibrant Selection in Measurement of Carbohydrate and Peptide Ion-Neutral Collision Cross Sections by Traveling Wave Ion Mobility Spectrometry**

Abby S. Gelb, Rebecca E. Jarratt, Yuting Huang, and Eric D. Dodds\*

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11403  DOI: 10.1021/ac503363m

**Intracellular Tracking of Single Native Molecules with Electroporation-Delivered Quantum Dots**

Chen Sun, Zhenning Cao, Min Wu, and Chang Lu\*

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11410 DOI: 10.1021/ac503365z

**Quencher-Free Fluorescent Method for Homogeneously Sensitive Detection of MicroRNAs in Human Lung Tissues**

Guichi Zhu, Li Liang, and Chun-yang Zhang\*

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11417  DOI: 10.1021/ac503407e

**Achieving Reproducible Performance of Electrochemical, Folding Aptamer-Based Sensors on Microelectrodes: Challenges and Prospects**

Juan Liu, Samiullah Wagan, Melissa Dávila Morris, James Taylor, and Ryan J. White\*

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11425  DOI: 10.1021/ac503522f

**Thermodynamics-based Rational Design of DNA Block Copolymers for Quantitative Detection of Single-Nucleotide Polymorphisms by Affinity Capillary Electrophoresis**

Ayumi Kimura, Naoki Kanayama, Atsushi Ogawa, Hideaki Shibata, Hideo Nakashita, Tohru Takarada,\* and Mizuo Maeda

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11434  DOI: 10.1021/ac503540q

**Target-Responsive DNzyme Cross-Linked Hydrogel for Visual Quantitative Detection of Lead**

Yishun Huang, Yanli Ma, Yahong Chen, Xuemeng Wu, Luting Fang, Zhi Zhu,\* and Chaoyong James Yang\*

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11440  DOI: 10.1021/ac503546r

**Poly  $\beta$ -Cyclodextrin/TPdye Nanomicelle-based Two-Photon Nanoprobe for Caspase-3 Activation Imaging in Live Cells and Tissues**

Huijuan Yan, Leiliang He, Wenjie Zhao, Jishan Li,\* Yue Xiao, Ronghua Yang,\* and Weihong Tan

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11451 DOI: 10.1021/ac503616d

**Low-Cost and Highly Sensitive Immunosensing Platform for Aflatoxins Using One-Step Competitive Displacement Reaction Mode and Portable Glucometer-Based Detection**

Dianping Tang,\* Youxiu Lin, Qian Zhou, Yuping Lin, Peiwu Li, Reinhard Niessner, and Dietmar Knopp\*

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11459  DOI: 10.1021/ac503622n

**Heterogeneous Nano Metal–Organic Framework Fluorescence Probe for Highly Selective and Sensitive Detection of Hydrogen Sulfide in Living Cells**

Yu Ma, Hao Su, Xuan Kuang, Xiangyuan Li, Tingting Zhang, and Bo Tang\*

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**Analysis of Binary Mixtures of Aqueous Aromatic Hydrocarbons with Low-Phase-Noise Shear-Horizontal Surface Acoustic Wave Sensors Using Multielectrode Transducer Designs**

Florian Bender, Rachel E. Mohler, Antonio J. Ricco, and Fabien Josse\*

**Additions and Corrections****Correction to VAMAS Interlaboratory Study for Desorption Electrospray Ionization Mass Spectrometry (DESI MS) Intensity Repeatability and Constancy**

Elzbieta Gurdak,\* Felicia M. Green, Paulina D. Rakowska,\* Martin P. Seah, Tara L. Salter, and Ian S. Gilmore