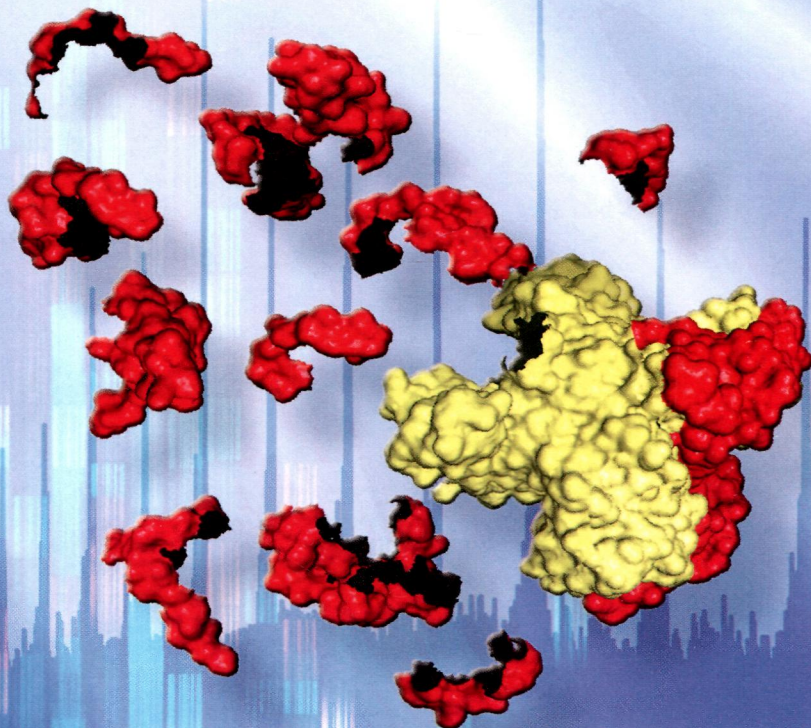


**Native Top-Down  
Electrospray Ionization-Mass  
Spectrometry of 158 kDa  
Protein Complex by  
High-Resolution  
Fourier Transform Ion  
Cyclotron Resonance  
Mass Spectrometry**



**ON THE COVER:** Native top-down mass spectrometry with electron capture dissociation coupled to high-resolution Fourier transform ion cyclotron resonance MS yields information on the surface topology of protein complexes. Image created by Huilin Li (UCLA), Valerie Cox (Bruker Daltonics), and Piriya Wongkongkathep (UCLA).

## Editors' Highlights

967  [dx.doi.org/10.1021/ac403753r](https://doi.org/10.1021/ac403753r)  
**Visualizing and Quantifying Protein PolySUMOylation at the Single-Molecule Level**  
Yong Yang and Chun-yang Zhang\*


## Letters to Analytical Chemistry


973  [dx.doi.org/10.1021/ac403478z](https://doi.org/10.1021/ac403478z)  
**Electrochemical Biosensing Platform Using Hydrogel Prepared from Ferrocene Modified Amino Acid as Highly Efficient Immobilization Matrix**  
Fengli Qu, Yi Zhang, Avraham Rasooly, and Minghui Yang\*

977  [dx.doi.org/10.1021/ac403777g](https://doi.org/10.1021/ac403777g)  
**Image-Based Single-Cell Sorting via Dual-Photopolymerized Microwell Arrays**  
Tao Sun, Joseph Kovac, and Joel Voldman\*

## Technical Notes

982  [dx.doi.org/10.1021/ac402823n](https://doi.org/10.1021/ac402823n)  
**Rapid Matrix-Assisted Laser Desorption/Ionization Time-of-Flight Mass Spectrometry Imaging with Scanning Desorption Laser Beam**  
Antonín Bednařík, Pavel Kuba, Eugene Moskovets, Iva Tomalová, Pavel Krásenský, Pavel Houška, and Jan Preisler\*

987  [dx.doi.org/10.1021/ac403261s](https://doi.org/10.1021/ac403261s)  
**Biomolecular Analysis and Biological Tissue Diagnostics by Electrospray Ionization with a Metal Wire Inserted Gel-Loading Tip**  
Mridul Kanti Mandal,\* Kentaro Yoshimura, Subhrakanti Saha, Zhan Yu, Sen Takeda, and Kenzo Hiraoka\*

993  dx.doi.org/10.1021/ac403438c  
**Speciation without Chromatography Using Selective Hydride Generation: Inorganic Arsenic in Rice and Samples of Marine Origin**  
Stanislav Musil, Ásta H. Pétursdóttir, Andrea Raab, Helga Gunnlaugsdóttir, Eva Krupp, and Jörg Feldmann\*

## Articles


1000  dx.doi.org/10.1021/ac400867b  
**High-Throughput Solvent Assisted Ionization *Inlet* for Use in Mass Spectrometry**  
Beixi Wang and Sarah Trimpin\*


1007 dx.doi.org/10.1021/ac401081s  
**Confocal Raman Characterization of Different Protein Desorption Behaviors from Chromatographic Particles**  
Yuewu Xiao,\* Thomas Stone, Wilson Moya, Paul Killian, and Thomas Herget


1016  dx.doi.org/10.1021/ac401523e  
**Enhancing the Detection Limit of Nanoscale Biosensors via Topographically Selective Functionalization**  
Mark A. Lifson, Dhrubajyoti Basu Roy, and Benjamin L. Miller\*

1023 dx.doi.org/10.1021/ac401751r  
**Failure of ESI Spectra to Represent Metal-Complex Solution Composition: A Study of Lanthanide–Carboxylate Complexes**  
Luther W. McDonald IV, James A. Campbell, and Sue B. Clark\*


1030  dx.doi.org/10.1021/ac401764c  
**Monitoring Aptamer–Protein Interactions Using Tunable Resistive Pulse Sensing**  
Emily R. Billinge, Murray Broom, and Mark Platt\*


1038  dx.doi.org/10.1021/ac401797m  
**Simple Nanoparticle-Based Luminometric Method for Molecular Weight Determination of Polymeric Compounds**  
Sari Pihlasalo,\* Maria Virtamo, Nicolas Legrand, Pekka Hänninen, and Harri Härnä


1045  dx.doi.org/10.1021/ac402230h  
***In Vivo* Proton–Electron Double-Resonance Imaging of Extracellular Tumor pH Using an Advanced Nitroxide Probe**  
Alexandre Samouilov, Olga V. Efimova, Andrey A. Bobko, Ziqi Sun, Sergey Petryakov, Timothy D. Eubank, Dmitrii G. Trofimov, Igor A. Kirilyuk, Igor A. Grigor'ev, Wataru Takahashi, Jay L. Zweier, and Valery V. Khrantsov\*

1053  dx.doi.org/10.1021/ac403019e  
**Ultrasensitive Apurinic/Apyrimidinic Endonuclease 1 Immunosensing Based on Self-Enhanced Electrochemiluminescence of a Ru(II) Complex**  
Ying Zhuo,\* Ni Liao, Ya-Qin Chai, Guo-Feng Gui, Min Zhao, Jing Han, Yun Xiang, and Ruo Yuan\*

1061 dx.doi.org/10.1021/ac402628t  
**Discrete Free-Surface Millifluidics for Rapid Capture and Analysis of Airborne Molecules Using Surface-Enhanced Raman Spectroscopy**  
Brian D. Piorek,\* Chrysafis Andreou, Martin Moskovits, and Carl D. Meinhardt

1067  dx.doi.org/10.1021/ac402603j  
**Examining the Interactions of the Splicing Factor MBNL1 with Target RNA Sequences via a Label-Free, Multiplex Method**  
Amrita R. Yadav, Charles R. Mace, and Benjamin L. Miller\*


1076  dx.doi.org/10.1021/ac4026384  
**Highly Sensitive and Specific Multiplexed MicroRNA Quantification Using Size-Coded Ligation Chain Reaction**  
Pengbo Zhang, Jiangyan Zhang, Chengli Wang, Chenghui Liu,\* Hui Wang, and Zhengping Li\*

1083  dx.doi.org/10.1021/ac4026576  
**Atomic Force Microscope Controlled Topographical Imaging and Proximal Probe Thermal Desorption/Ionization Mass Spectrometry Imaging**  
Olga S. Ovchinnikova, Kevin Kjoller, Gregory B. Hurst, Dale A. Pelletier, and Gary J. Van Berkel\*

1091 dx.doi.org/10.1021/ac402753j  
**Quantitative Compositional Profiling of Conjugated Quantum Dots with Single Atomic Layer Depth Resolution via Time-of-Flight Medium-Energy Ion Scattering Spectroscopy**  
Kang-Won Jung, Hyunung Yu, Won Ja Min, Kyu-Sang Yu, M. A. Sortica, Pedro L. Grande, and DaeWon Moon\*

1098  dx.doi.org/10.1021/ac402869b  
**Integrated Analysis of Seaweed Components during Seasonal Fluctuation by Data Mining Across Heterogeneous Chemical Measurements with Network Visualization**  
Kengo Ito, Kenji Sakata, Yasuhiro Date, and Jun Kikuchi\*

1106 dx.doi.org/10.1021/ac403621c  
**Monitoring of Human Chemical Signatures Using Membrane Inlet Mass Spectrometry**  
Stamatios Giannoukos, Boris Brkić,\* Stephen Taylor,\* and Neil France

1115  dx.doi.org/10.1021/ac4028774  
**Sensitive Chemiluminescence Immunoassay for *E. coli* O157:H7 Detection with Signal Dual-Amplification Using Glucose Oxidase and Laccase**  
Yun Zhang, Chen Tan, Ruihua Fei, Xiaoxiao Liu, Yuan Zhou, Jing Chen, Huanchun Chen, Rui Zhou, and Yonggang Hu\*

1123  dx.doi.org/10.1021/ac402894z  
**Fluorescence “Turn On” Detection of Mercuric Ion Based on Bis(dithiocarbamate)copper(II) Complex Functionalized Carbon Nanodots**  
Chao Yuan, Bianhua Liu,\* Fei Liu, Ming-Yong Han, and Zhongping Zhang\*



1131  dx.doi.org/10.1021/ac4029054

**Enhancing the Analytical Performance of Electrochemical RNA Aptamer-Based Sensors for Sensitive Detection of Aminoglycoside Antibiotics**

Lauren R. Schoukroun-Barnes, Samuillah Wagan, and Ryan J. White\*

1138  dx.doi.org/10.1021/ac403067w

**Robust Method for Investigating Nitrogen Metabolism of <sup>15</sup>N Labeled Amino Acids Using AccQ•Tag Ultra Performance Liquid Chromatography-Photodiode Array-Electrospray Ionization-Mass Spectrometry: Application to a Parasitic Plant-Plant Interaction**

Zachary Gaudin, Delphine Cerveau, Nathalie Marnet, Alain Bouchereau, Philippe Delavault, Philippe Simier, and Jean-Bernard Pouvreau\*

1146  dx.doi.org/10.1021/ac403146y

**Highly Flexible UV-Vis Radiation Sources and Novel Detection Schemes for Spectrophotometric HPLC Detection**

Karsten G. Kraiczek,\* R. Bonjour, Y. Salvadé, and Roland Zengerle

1153 dx.doi.org/10.1021/ac402997e

**Kläui Ligand Thin Films for Rapid Plutonium Analysis by Alpha Spectrometry**

Susan K. Hanson,\* Alexander H. Mueller, and Warren J. Oldham Jr.\*

1160  dx.doi.org/10.1021/ac403110u

**Focus: A Robust Workflow for One-Dimensional NMR Spectral Analysis**

Arnald Alonso, Miguel A. Rodríguez, María Vinaixa, Raül Tortosa, Xavier Correig, Antonio Julià,\* and Sara Marsal

1170  dx.doi.org/10.1021/ac403060d

**Trypsin-Catalyzed N-Terminal Labeling of Peptides with Stable Isotope-Coded Affinity Tags for Proteome Analysis**

Yanbo Pan, Mingliang Ye,\* Hao Zheng, Kai Cheng, Zhen Sun, Fangjie Liu, Jing Liu, Keyun Wang, Hongqiang Qin, and Hanfa Zou\*

1178  dx.doi.org/10.1021/ac4032675

**Self-Catalytic Growth of Unmodified Gold Nanoparticles as Conductive Bridges Mediated Gap-Electrical Signal Transduction for DNA Hybridization Detection**

Jing Zhang, Huagui Nie,\* Zhan Wu, Zhi Yang, Lijie Zhang, Xiangju Xu, and Shaoming Huang\*

1186  dx.doi.org/10.1021/ac403352e

**Development and Validation of a Sensitive UPLC-ESI-MS/MS Method for the Simultaneous Quantification of 15**

**Endocannabinoids and Related Compounds in Milk and Other Biofluids**

Sandra Gouveia-Figueira and Malin L. Nording\*

1196  dx.doi.org/10.1021/ac403347e


**Detecting Plasmon Resonance Energy Transfer with Differential Interference Contrast Microscopy**

Ashley E. Augspurger, Anthony S. Stender, Rui Han, and Ning Fang\*

1202  dx.doi.org/10.1021/ac403385y

**Comparison of Information-Dependent Acquisition, SWATH, and MS<sup>AM</sup> Techniques in Metabolite Identification Study Employing Ultrahigh-Performance Liquid Chromatography-Quadrupole Time-of-Flight Mass Spectrometry**

Xiaochun Zhu,\* Yuping Chen, and Raju Subramanian

1210  dx.doi.org/10.1021/ac403386q

**Isomerization Kinetics of AT Hook Decapeptide Solution Structures**

Emily R. Schenk, Mark E. Ridgeway, Melvin A. Park, Fenfei Leng, and Francisco Fernandez-Lima\*

1215  dx.doi.org/10.1021/ac4035696

**Progress in Detection and Structural Characterization of Glycosphingolipids in Crude Lipid Extracts by Enzymatic Phospholipid Disintegration Combined with Thin-Layer Chromatography Immunodetection and IR-MALDI Mass Spectrometry**

Ivan U. Kouzel, Alexander Pirkel, Gottfried Pohlentz, Jens Soltwisch, Klaus Dreisewerd, Helge Karch, and Johannes Müthing\*

1223  dx.doi.org/10.1021/ac403536a


**Encapsulation of Strongly Fluorescent Carbon Quantum Dots in Metal-Organic Frameworks for Enhancing Chemical Sensing**

Xiaomei Lin, Gongmin Gao, Liyan Zheng, Yuwu Chi,\* and Guonan Chen

1229  dx.doi.org/10.1021/ac403541a

**Investigation of Gender-Specific Exhaled Breath Volatome in Humans by GCxGC-TOF-MS**

Mrinal Kumar Das, Subasa Chandra Bishwal, Aleena Das, Deepti Dabral, Ankur Varshney, Vinod Kumar Badireddy, and Ranjan Nanda\*

1238  dx.doi.org/10.1021/ac4035532

**Identification of Allosteric Nucleotide Sites of Tetramethylrhodamine-Labeled Aptamer for Noncompetitive Aptamer-Based Fluorescence Anisotropy Detection of a Small Molecule, Ochratoxin A**

Qiang Zhao,\* Qin Lv, and Hailin Wang

1246  dx.doi.org/10.1021/ac403672u

**Fully-Automated Approach for Online Dried Blood Spot Extraction and Bioanalysis by Two-Dimensional-Liquid Chromatography Coupled with High-Resolution Quadrupole Time-of-Flight Mass Spectrometry**

Regina V. Oliveira, Jack Henion,\* and Enaksha Wickremsinhe

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[dx.doi.org/10.1021/ac4035949](https://doi.org/10.1021/ac4035949)


**Analysis of Covalent Modifications of Proteins by Oxidized Phospholipids Using a Novel Method of Peptide Enrichment**  
Detao Gao, Belinda Willard, and Eugene A. Podrez\*

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[dx.doi.org/10.1021/ac403616c](https://doi.org/10.1021/ac403616c)

**Superior Fluorescent Probe for Detection of Cardiolipin**

Chris W. T. Leung, Yuning Hong, Jonas Hanske, Engui Zhao, Sijie Chen, Ekaterina V. Pletneva,\* and Ben Zhong Tang\*

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[dx.doi.org/10.1021/ac403630a](https://doi.org/10.1021/ac403630a)

**MACRO: A Combined Microchip-PCR and Microarray System for High-Throughput Monitoring of Genetically Modified Organisms**


Ning Shao, Shi-Meng Jiang, Miao Zhang, Jing Wang, Shu-Juan Guo, Yang Li, He-Wei Jiang, Cheng-Xi Liu, Da-Bing Zhang, Li-Tao Yang,\* and Sheng-Ce Tao\*

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[dx.doi.org/10.1021/ac403674p](https://doi.org/10.1021/ac403674p)

**High-Performance Liquid Chromatographic Enantioseparation of Racemic Drugs Based on Homochiral Metal–Organic Framework**

Xuan Kuang, Yu Ma, Hao Su, Jine Zhang, Yu-Bin Dong,\* and Bo Tang\*

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[dx.doi.org/10.1021/ac403686a](https://doi.org/10.1021/ac403686a)

**Enantiomeric Separations of Chiral Sulfonic and Phosphoric Acids with Barium-Doped Cyclofructan Selectors via an Ion Interaction Mechanism**


Jonathan P. Smuts, Xin-Qi Hao, Zhaobin Han, Curran Parpia, Michael J. Krische, and Daniel W. Armstrong\*

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[dx.doi.org/10.1021/ac403705c](https://doi.org/10.1021/ac403705c)

**Ex Vivo Chemical Cytometric Analysis of Protein Tyrosine Phosphatase Activity in Single Human Airway Epithelial Cells**

Ryan M. Phillips, Lisa A. Dailey, Eric Bair, James M. Samet, and Nancy L. Allbritton\*

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[dx.doi.org/10.1021/ac4038976](https://doi.org/10.1021/ac4038976)

**Extracting Kinetics from Affinity Capillary Electrophoresis (ACE) Data: A New Blade for the Old Tool**


Mirzo Kanoatov, Leonid T. Cherney, and Sergey N. Krylov\*

1306 

[dx.doi.org/10.1021/ac404121c](https://doi.org/10.1021/ac404121c)

**In Situ-Generated Nano-Gold Plasmon-Enhanced Photoelectrochemical Aptasensing Based on Carboxylated Perylene-Functionalized Graphene**

Jing Li, Wenwen Tu, Hongbo Li,\* Min Han, Yaqian Lan, Zhihui Dai,\* and Jianchun Bao

 Supporting Information available via online article