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**Raman spectroscopy of the iron-based superconductors**[1]

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The multiband nature of iron-based superconductors (FeSCs) give rise to a rich temperature-doping phase diagram of competing orders. The parent materials are antiferromagnetic metals. At low doping concentrations, a tetragonal to orthorhombic structure phase transition which breaks the four-fold rotational symmetry is closely followed by the formation of a spin-density wave order which breaks the translational symmetry by doubling the unit cell. The electronic nematic phase forms when the discrete four-fold rotational symmetry is broken while the translational symmetry remains. Superconductivity often emerges in close proximity to the nematic phase. The system provides a platform to study coexistence or competition between the nematic order, the density-wave order and superconductivity. Here we provide a comprehensive polarization resolved electronic Raman spectroscopy study of the intertwined phases in two systems of FeSCs: $A\mathrm{Fe_2As_2}$($A$=Ba,Ca,Eu) and $\mathrm{FeSe_{1-x}S_x}$. Critical enhancement of the nematic charge fluctuations is observed in the non-symmetric Raman response above the structure phase transition temperature in $A\mathrm{Fe_2As_2}$ and $\mathrm{FeSe_{1-x}S_x}$. The charge fluctuations are interpreted in terms of inter-orbital charge quadrupole excitations. We compare the susceptibilities measured by Raman scattering, elastic shear modulus $\mathrm{C_{66}}$, elastoresistance $\mathrm{m_{66}}$, and nuclear quadrupole resonance (NQR), and demonstrate the universality of the susceptibilities measured by these probes. Anisotropic gap opening in the Raman response is observed in the spin-density wave phase of $A\mathrm{Fe_2As_2}$. In the non-magnetic $\mathrm{FeSe_{1-x}S_x}$, we discover a gap in the Raman response, which is similar to the observations in $A\mathrm{Fe_2As_2}$. The data in $\mathrm{FeSe_{1-x}S_x}$ suggests a stripe type charge quadrupole order in the orthorhombic phase, which could result in strongly anisotropic electronic properties and orbital dependent superconductivity, as observed in ARPES and STM experiments. Research at Rutgers was done in collaboration with G. Blumberg and S.-F. Wu.

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