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| **Title** | **Ground-State Depletion Microscopy: Detection Sensitivity of Single-Molecule Optical Absorption at Room Temperature** |
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| **Abstract** | Optical studies of single molecules in ambient environments, which have led to broad applications, are primarily based on fluorescence detection. Direct detection of optical absorption with single-molecule sensitivity at room temperature is difficult because absorption is not a background-free measurement and is often complicated by sample scattering. Here we report ground-state depletion microscopy for ultrasensitive detection of absorption contrast. We image 20 nm gold nanoparticles as an initial demonstration of this microscopy. We then demonstrate the detection of an absorption signal from a single chromophore molecule at room temperature. This is accomplished by using two tightly focused collinear continuous-wave laser beams at different wavelengths, both within a molecular absorption band, one of which is intensity modulated at a high frequency (>MHz). The transmission of the other beam is found to be modulated at the same frequency due to ground state depletion. The signal of single chromophore molecules scanned across the common laser foci can be detected with shot-noise limited sensitivity. This measurement represents the ultimate detection sensitivity of nonlinear optical spectroscopy at room temperature. |
| **Laser Quantum Product** | Ventus 532nm |
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