Longitudinal Field Modes Probed by Single Molecules

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Longitudinal Field Modes Probed by Single Molecules

- measurement of field distribution near focus of strongly focused radially polarized field mode
- probe: molecules with fixed absorption dipole orientation

and vice versa

L. Novotny, M.R. Beversluis, K.S. Youngworth, and T.G. Brown
PRL 86, 5251 (2001)

Experimental setup

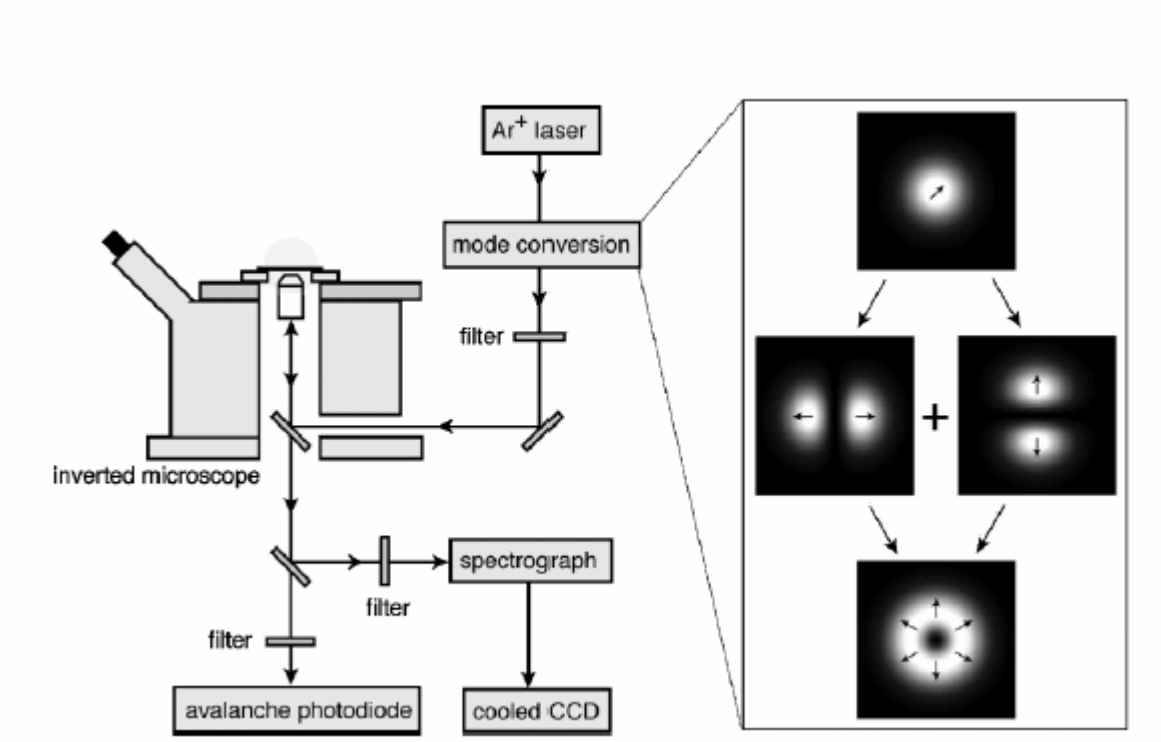

FIG. 1. Experimental setup: The fundamental laser mode of an Ar^+ laser ($\lambda = 488$ nm) is converted into a radially polarized mode and focused on a dielectic sample consisting of a PMMA layer with embedded dye molecules. The strength of the longitudinal field at the laser focus is determined by scanning individual molecules with fixed absorption dipole orientation through the laser focus and monitoring the fluorescence emission rate.

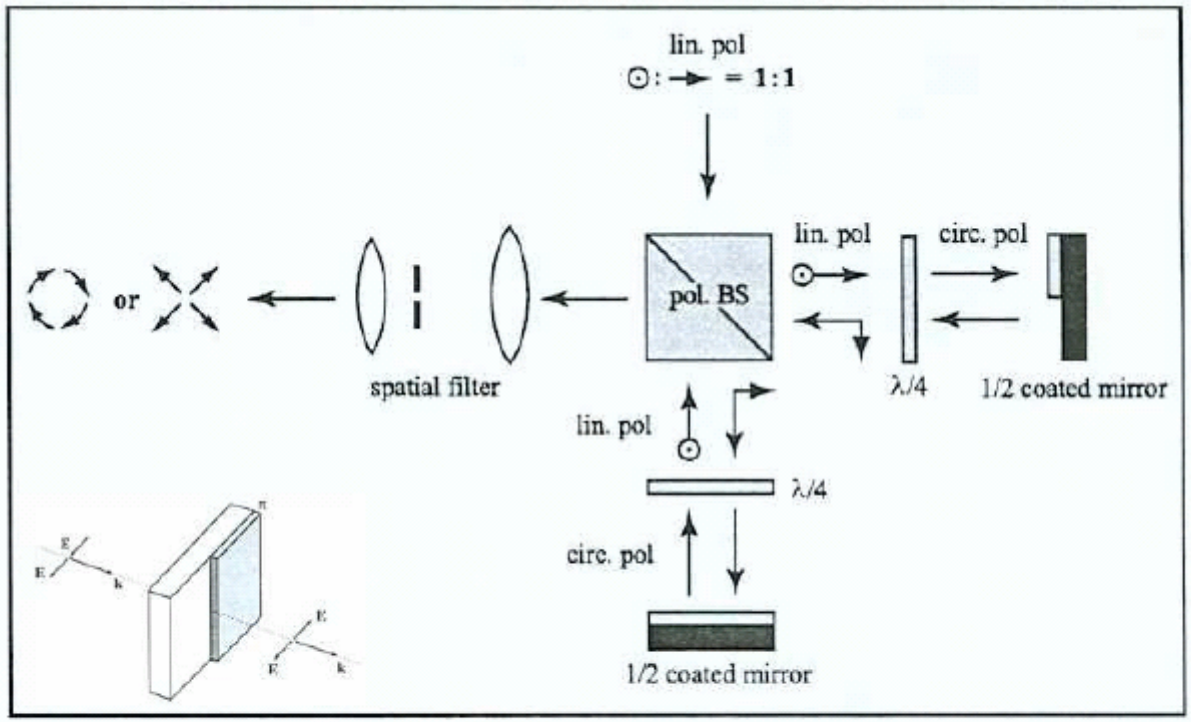

Figure 17: Simple setup to produce a Doughnut-mode. RP or AP polarization are controlled by the position of the mirrors. The inset shows a detail of the half coated mirror. From [Novotny and Hecht, "Principles of Nano-Optics"]

Calculations

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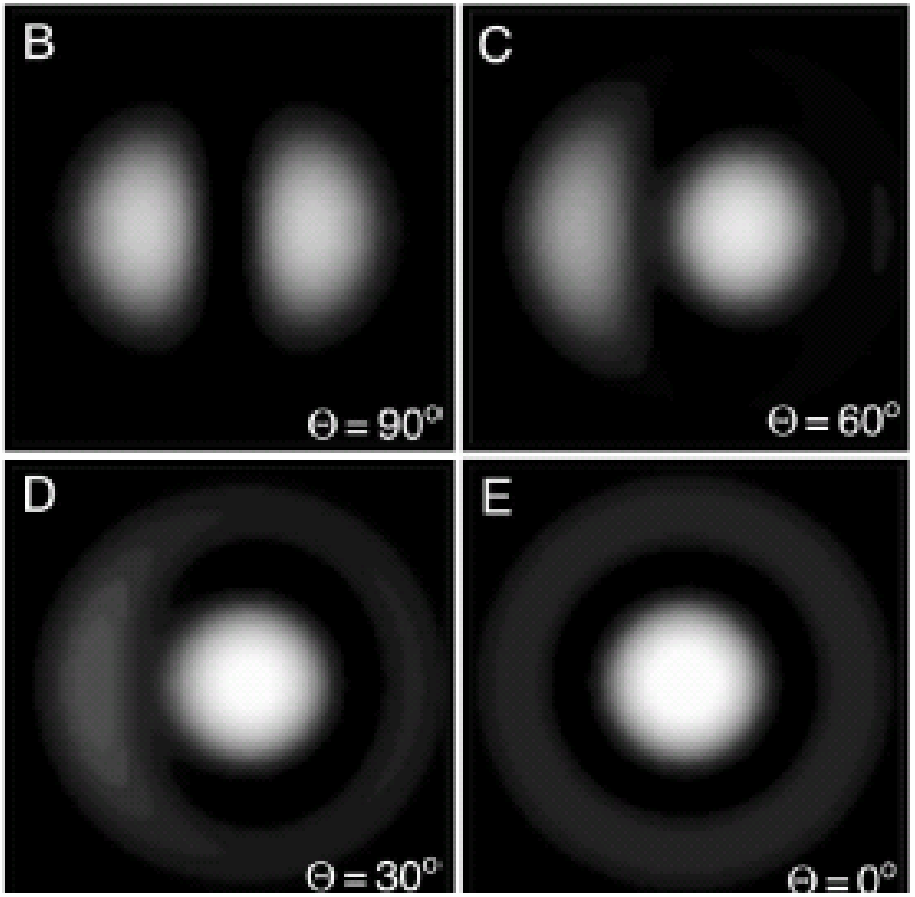

FIG. 2. (A) Comparison of longitudinal field strength E_z^2, transverse field strength E_ρ^2, and total field strength E^2 as a function of radial coordinate, evaluated 2 nm beneath the interface. (B)–(E) Calculated emission rate images $[R(x, y)]$ for different polar orientations of a molecule in the x-z plane. θ denotes the angle between the dipole axis and the beam axis.

Confirmation

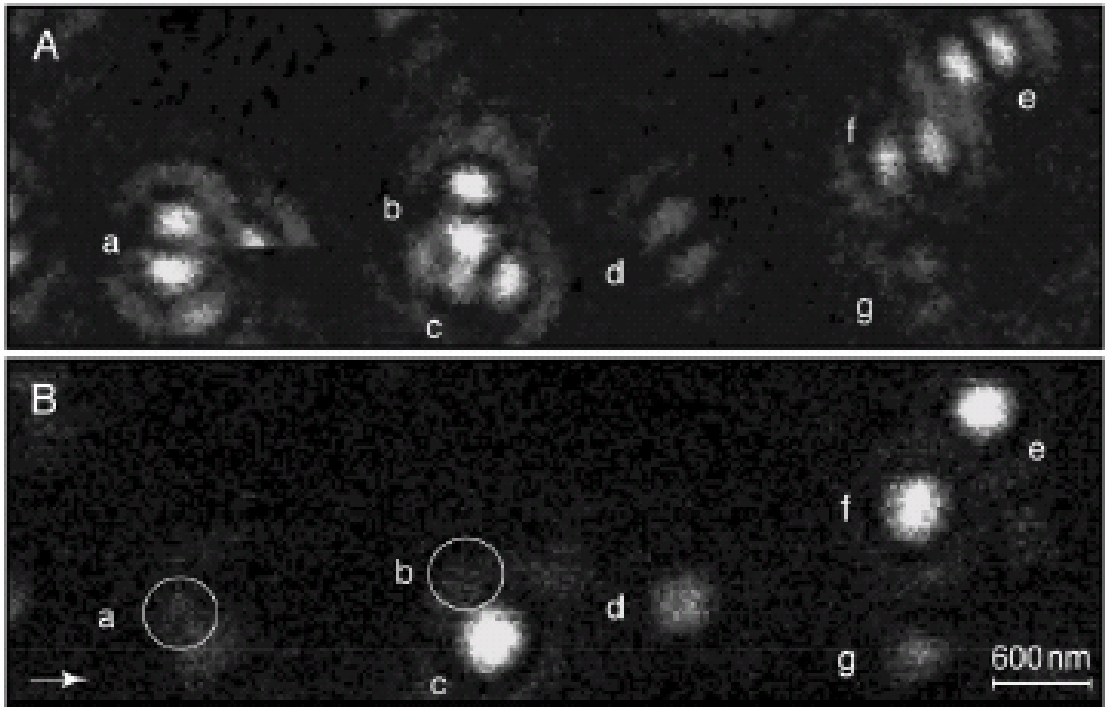

FIG. 3. Fluorescence rate images $[R(x, y)]$ of molecules oriented transverse to the beam axis. (A) Excitation with a focused radially polarized beam. Adjacent lobes define the direction of the molecular absorption dipole (cf. Fig. 2B). (B) Excitation with a focused fundamental Gaussian beam polarized in the direction of the arrow. Molecules a and b are not excited because their absorption dipole is perpendicular to the polarization.

Determination of orientation of molecular absorption dipole

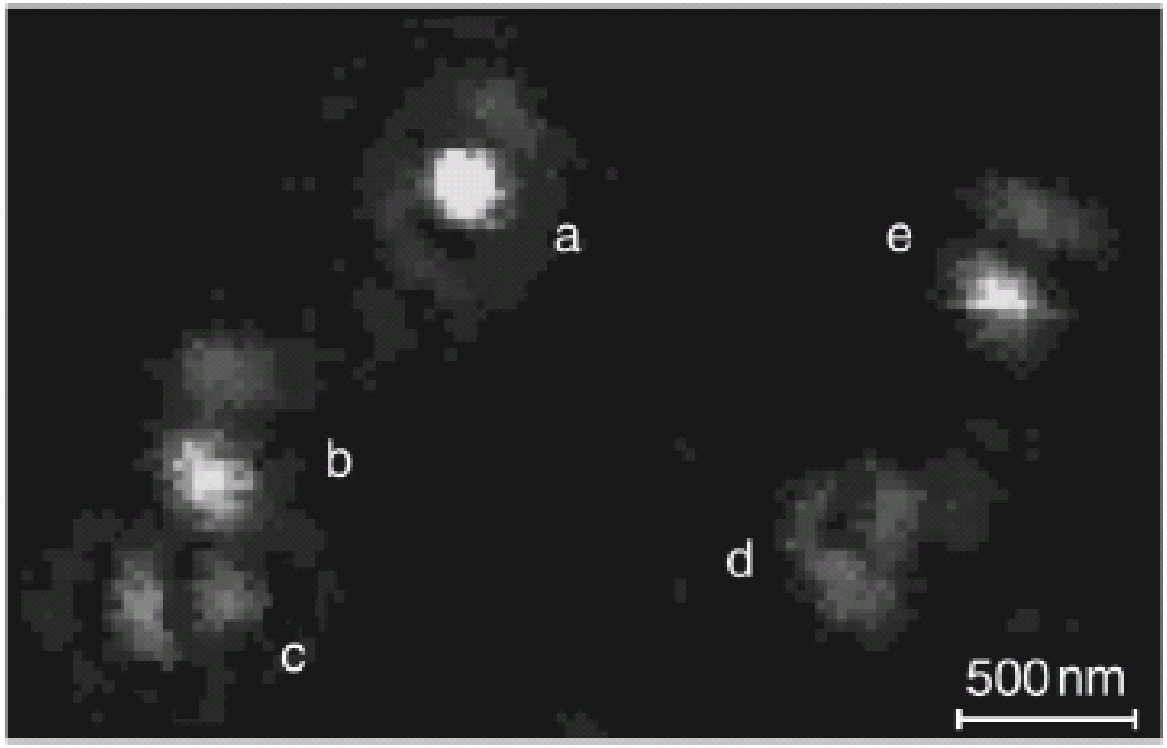

FIG. 4. Fluorescence rate images $[R(x, y)]$ of arbitrarily oriented molecules excited with a focused radially polarized beam. The dipole moment of molecule a is nearly longitudinal (cf. Fig. 2D), whereas the dipole moments of molecules c and d are transverse. Molecules b and e have an orientation of $\theta \approx 60°$ (cf. Fig. 2C).

Calculations

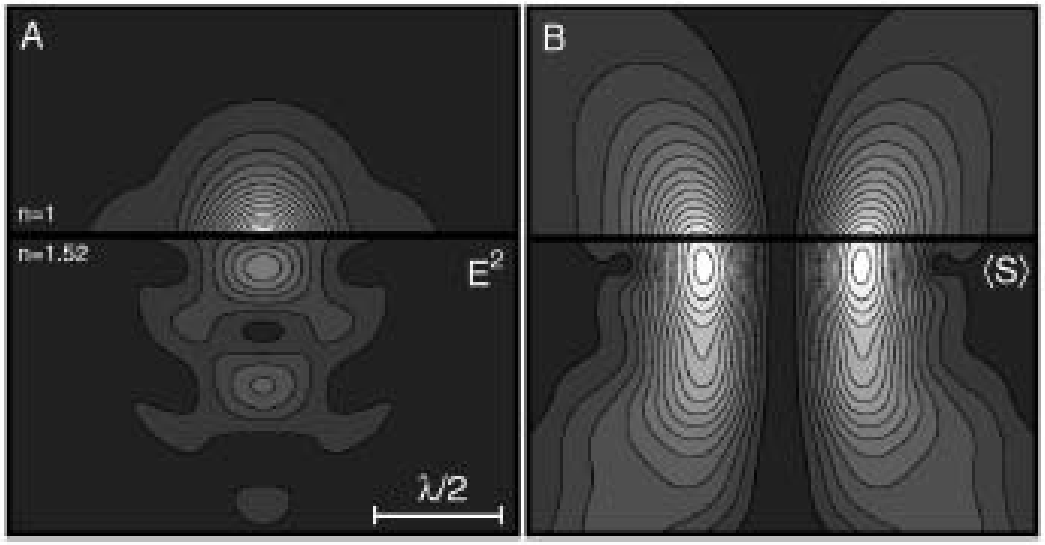

FIG. 5. (A) Electric field strength E^2 and (B) absolute value of the time averaged Poynting vector $\langle S \rangle$ of a radially polarized beam focused upward on the surface of a dielectric medium ($n = 1.52$). The longitudinal field E_z^2 centered on the beam axis does not propagate. Energy transport is associated only with the transverse field E_ρ^2.

Orientational Imaging of Single Molecules by Annular Illumination

- Imaging of orientation
- Scanning confocal microscope
- Excitation by focused annular beam

→ enhanced longitudinal electric field

B. Sick and B. Hecht, PRL 85, 4482 (2000)

Setup

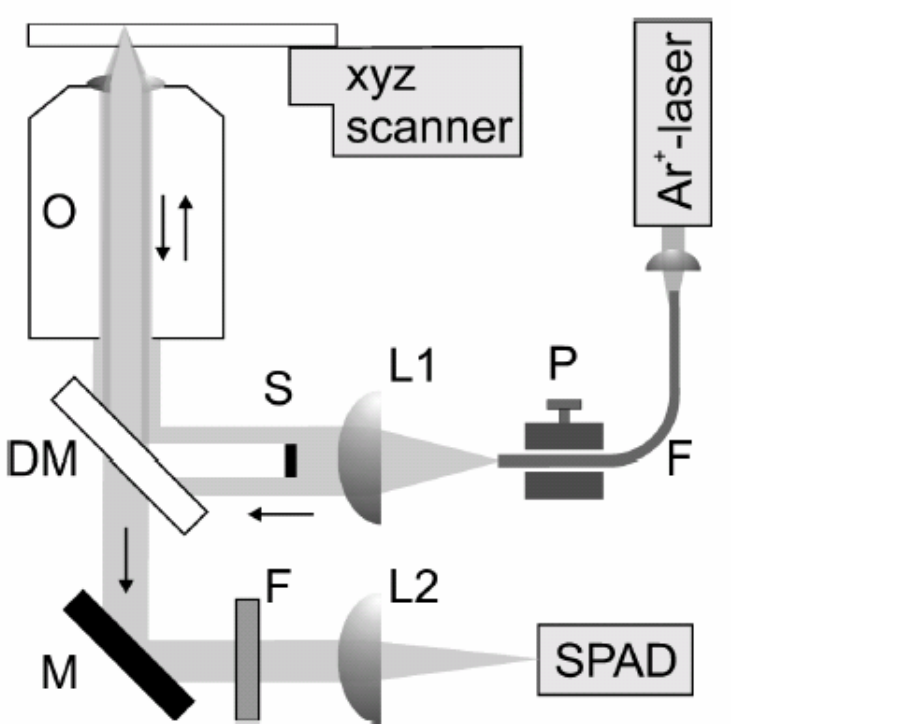

FIG. 1. Setup for annular illumination scanning confocal optical microscopy. L1: collimation lens ($f = 50$ mm); O: microscope objective (Leica, PL Fluotar, $\times 100$, 1.3 numerical aperture, ∞); DM: dichroic mirror; M: mirror; F: holographic notch filter; L2: focusing lens ($f = 300$ mm); SPAD: single photon counting avalanche photodiode (EG&G SPCM 200); S: obstruction disk (3 mm diam); P: polarization controller (Polarite, L.O.T.); F single mode fiber.

Typical image

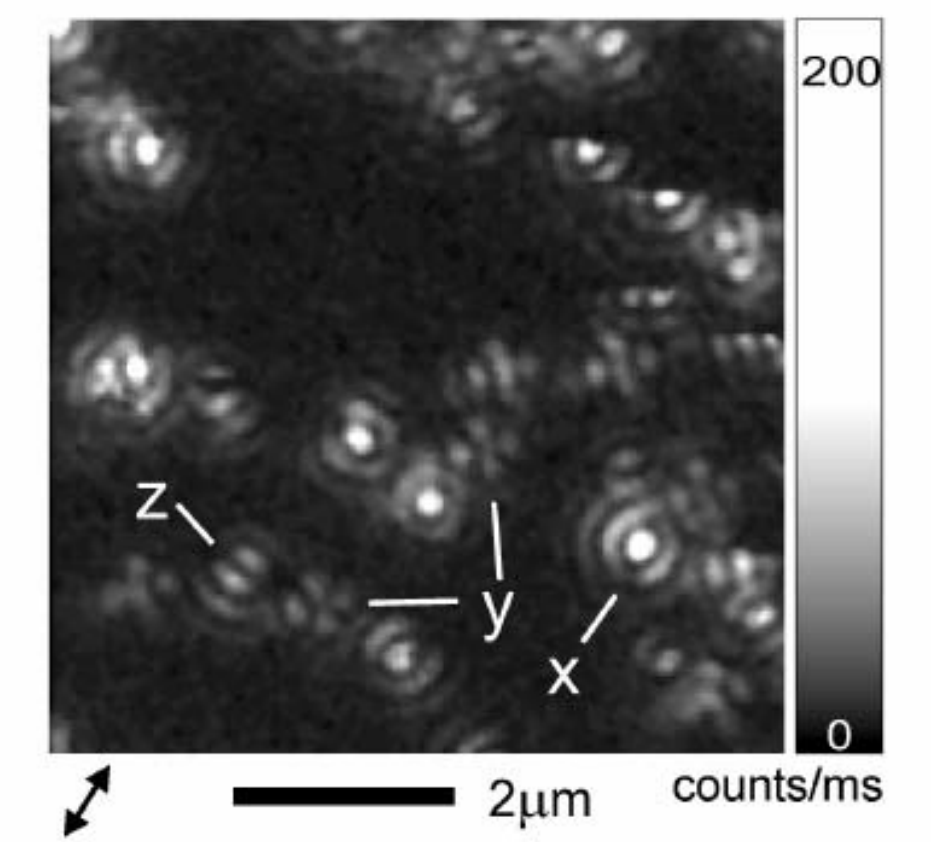

FIG. 2. Typical confocal single-molecule fluorescence image of DiI in a thin film of PMMA obtained with linearly polarized (arrow), annular illumination. The labeled patterns result from molecules that map the squared x, y, and z components of the electric field in the focus, respectively.

Calculated fluorescence patterns

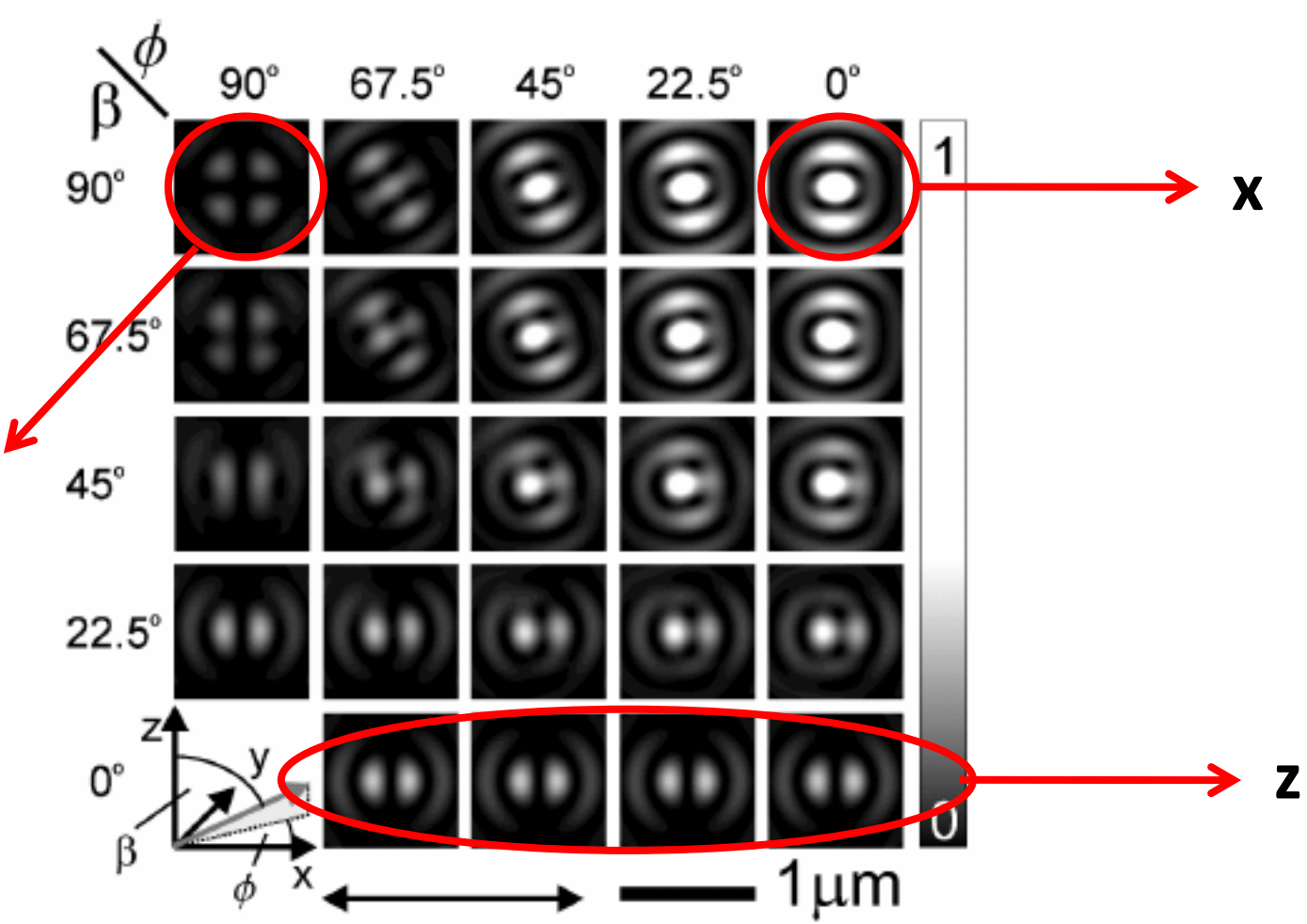

FIG. 4. Single-molecule fluorescence patterns 5 nm below the interface calculated from the field distribution according to (1). Coordinate system: definition of the dipole orientation angles β and ϕ with respect to the excitation polarization (double arrow).

$$R(\vec{\mathbf{r}}) = c|\hat{\mathbf{d}} \cdot \vec{\mathbf{E}}(\vec{\mathbf{r}})|^2,$$

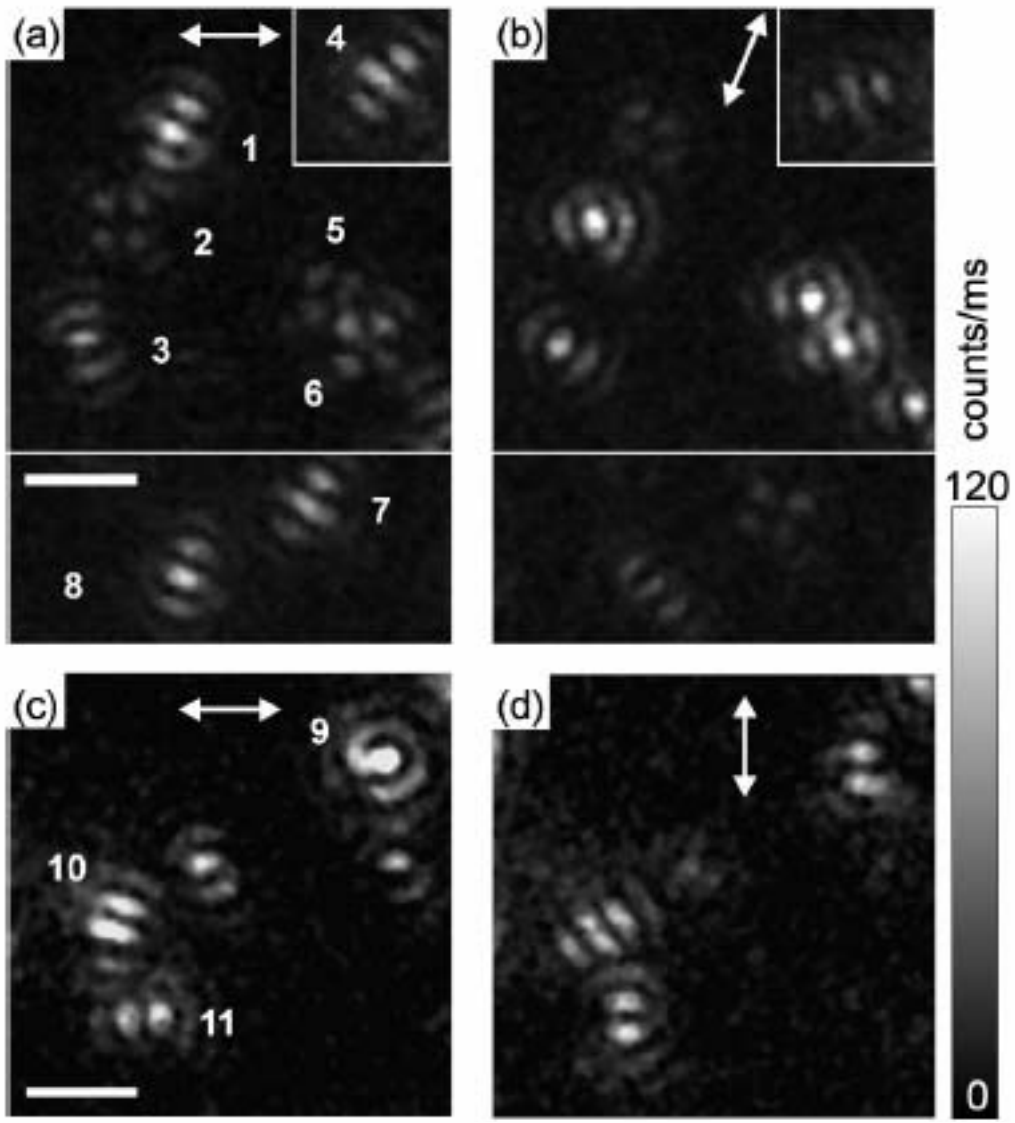

TABLE II. Orientation of the absorption dipole moment of single molecules, marked by numbers in Fig. 5, with respect to the polarization direction. Definition of ϕ and β as in Fig. 4.

Molecule	1	2	3	4	5	6	7	8	9	10	11
ϕ[°]	−45	90	45	−67.5	±90	90	−67.5	−30	45	−22.5	···
β[°]	67.5	67.5	90	90	90	67.5	90	90	−45	90	0

Summary

- probing of orientation of single molecules possible
- longitudinal mode more than twice as intense as transverse mode
- longitudinal mode does not transport energy

Thank you for your attention!