Probing the Magnetic Field of Light at Optical Frequencies

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Light is an electromagnetic wave composed of oscillating electric and magnetic fields, the one never occurring without the other. In light-matter interactions at optical frequencies, the magnetic component of light generally plays a negligible role. When we "see" or detect light, only its electric field is perceived; we are practically blind to its magnetic component. We used concepts from the field of metamaterials to probe the magnetic field of light with an engineered near-field aperture probe. We visualized with subwavelength resolution the magnetic- and electric-field distribution of propagating light.

As light interacts with matter, the force exerted by the electric field on a charge is $\mathcal{E}/\mathcal{E}$ larger than the force applied by the magnetic field, where $\mathcal{E}$ is the velocity of the charge and $\mathcal{E}$ is the speed of light. As a result, the response of a material to a magnetic field—its magnetic susceptibility—is a factor $10^{-7}$ smaller than the ease with which it is polarized, the dielectric susceptibility ($\varepsilon$). Only when the charges move extremely fast, such as in relativistic plasmas ($\varepsilon$, 3), can the magnetic and electric coupling become comparable. In atomic systems, even though the magnetic dipole coupling is extremely weak, it is important for fundamental tests of the standard model of particle physics ($\phi$). Magnetic light-matter interaction has been accomplished using artificial “magnetic” atoms. By tailoring the geometry of such subwavelength metallo-dielectric structures (so-called metamaterials), effective magnetic coupling is achievable in the microwave regime ($\phi$, 6). Photonic nanostructures that resonantly respond to the magnetic field at optical frequencies can now be fabricated ($\phi$–11). This magnetic resonance can be exploited to study fascinating phenomena, such as negative index of refraction ($\phi$), super-lensing (12), and cloaking ($\phi$, 14). Whereas many advances have been made in the control of light-matter coupling by magnetic means, the possibility of directly probing the magnetic field at optical frequencies has not yet been explored. The ability to directly probe the magnetic field of light would of course be beneficial to studies on metamaterials.

We used a near-field aperture probe, designed following split-ring resonator concepts, to detect the magnetic field at optical frequencies. Our probe was used to map the amplitude and phase of the magnetic field of propagating light. By simultaneously measuring the electric field as well, both constituent components of light (that is, the magnetic and the electric) can be mapped with subwavelength resolution.

We fabricated a nanostructured metallo-dielectric probe to detect the magnetic field at optical frequencies. A subwavelength aperture was created at the end of a tapered aluminum-coated single-mode fiber by means of focused ion-beam milling (15). An air gap of 40 nm was opened with focused ion-beam milling in the coating. Such a split-probe is shown in the lower image of Fig. 1B. We compare the optical signals measured with a split-probe with those measured with a standard, cylindrically symmetric, coated probe (Fig. 1B, top) that was used in the past (16, 17). To ensure that we probed the magnetic field rather than some signal caused by other electric-field components, a well-characterized single-mode Si$_3$N$_4$ ridge waveguide was used as a test structure (18, 19).

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the probed fields were obtained. The high symmetry of the standard probe can be used to distinguish the two in-plane components of the electric field of propagating light (17). As a result of the symmetry of the aperture, the in-plane components of the electric field couple to the orthogonal modes of the probe fiber and are detected in the two separate channels of the microscope (Ch1 and Ch2). For a formal description of what the probe measures, we refer to the supporting online material (SOM) (18). To probe the magnetic field of light, the magnetic field needs to be converted to an electric field before it can be measured at the detector. To achieve this conversion, the symmetry of the conventional probe needs to be broken. In the split-ring probe used in this work, this symmetry was broken by introducing a single gap in the side of the metal coating. This is crucial because a full ring or a ring with two opposing gaps would not exhibit the optical bia
erotropy required for converting the magnetic to an electric field (23, 24). Because the width of the air gap is much smaller than the wavelength of light inside the waveguide, the sensitivity to gradients in the electric field (7) is strongly suppressed.

The waveguide was first characterized by the performance of near-field measurements with a standard probe (Fig. 2A). Light propagates along \( \hat{x} \), the electric field is along \( \hat{y} \), and the magnetic field is along \( \hat{z} \). Because the waveguide contains only a weakly guided TE mode, the longitudinal component of the electric field is negligible (18, 25). The probe can be considered as a subwavelength metallic ring parallel to the sample surface because of its cylindrical symmetry and the extension of the evanescent fields in air of only \( \pm 100 \text{ nm} \) (25). The evanescent electric-field component \( E_y \) induces a dipolar charge distribution in the probe (SOM text). This induced oscillating dipole moment \( p_y \) couples to a propagating mode in the probe fiber and can be detected at the other end of the fiber. The magnetic field \( B_z \) instead generates a circular current in the ring, as described by Faraday’s law. As a result, the probe exhibits a magnetic dipole moment \( m_y \) in analogy with the magnetic response of a metallic cylinder (5). However, the radiation pattern of the magnetic dipole lies in the \( xy \) plane, and the cylindrical symmetry of the system forbids coupling of this magnetic dipole to the propagating modes in the probe fiber that propagate along \( \hat{z} \). Therefore, the fields detected with a standard probe are the in-plane electric fields (17, 26).

Figure 2B shows the line traces of the amplitude detected on Ch1 (red) and Ch2 (green), which were obtained by scanning the probe along the center of the waveguide. Because the only nonvanishing component of the electric field is along \( \hat{y} \), we can attribute the signals detected by Ch1 and Ch2 to \( E_x \) and \( E_y \), respectively (Fig. 2A). Because in the center of the waveguide the longitudinal component (\( E_z \)) vanishes (25), Fig. 2B allows us to infer the experimental extinction ratio of the two polarization channels. Because the ratio between the two amplitude signals is \( \pm 1/20 \) (in terms of intensity, \( \pm 1/400 \)), we were able to separate the two polarization states. As expected, a clear spatial amplitude modulation is evident in both channels. Because the spatial period (\( \approx 500 \text{ nm} \)) is half of a wavelength in the waveguide (18), this modulation is attributed to the standing wave caused by the interference between the forward-propagating light and the small fraction of light that is reflected by the end-facet of the waveguide. The maxima of the amplitude modulation in Ch1 and Ch2 are not shifted in space because both channels probe electric fields.

The sensitivity of the probe to the various field components of light changes drastically when the split probe is used. The air gap is oriented along \( \hat{y} \) (Fig. 2C). In analogy to the standard probe, \( E_x \) and \( B_z \) will induce in the split probe an electric and magnetic dipole moment \( p_y \) and \( m_y \), respectively. Similar to the cylindrical probe, the dipole moment \( p_y \) will generate an optical signal in Ch1. However, because of the air gap the magnetically induced current cannot flow completely around the ring and will produce a time-varying dipolar charge distribution across the gap. This in-plane magnetically induced electric dipole moment can now couple to the fiber of the probe. Because the polarization of this radiation is along \( \hat{x} \), the signal corresponding to \( B_z \) will be detected by Ch2. Hence, the optical signals with an electric and magnetic origin are detected in Ch1 and Ch2, respectively.

Figure 2D shows the measured amplitude of the Ch1 and Ch2 signals obtained with a split probe. In contrast to Fig. 2B, the signals in the two channels now have comparable magnitude. The standing wave induced amplitude modulation of the two signals is roughly equal. However, the most important difference between Fig. 2B and 2D is that the maxima of the two standing waves are shifted in space by half a period. It is well known that in a standing light wave, the amplitude of the magnetic field is shifted in space by half of a period with respect to the amplitude of the electric field (20). The spatial shift of the local maxima in Fig. 2D is therefore a clear signature that Ch2 detects the light generated by the coupling with \( B_z \). Thus, we have probed the out-of-plane component of the magnetic field.

To verify our claim of magnetic sensitivity of the probe, we performed an additional check. The same split-probe was used to measure on a waveguide oriented along \( \hat{y} \) while keeping the air gap oriented along \( \hat{y} \) (Fig. 2E). In this configuration, the electric field is along \( \hat{x} \) rather than \( \hat{y} \), and thus it should be detected by Ch2. However, \( B_z \) should also be probed by Ch2 because the orientations of the probe and, consequently, of the magnetically induced electric dipole moment have not changed. This means that the channel with the higher signal should now be Ch2. This was indeed experimentally observed, as shown in Fig. 2F. Although the ratio between the Ch1 and Ch2 signals is only \( \approx 0.27 \) (in terms of intensity, \( \approx 0.07 \)), it was higher than expected (see Fig. 2B). We attribute this to a minute in-plane rotation of the air gap with respect to \( \hat{y} \). When the air-gap is not perfectly aligned with \( \hat{y} \), the probe projects a fraction of \( B_z \) on Ch1. More important, because in this configuration the split probe does not separate \( E_x \) and \( B_z \), the amplitude maxima of the two channels are no.

**Fig. 1.** (A) Schematic of the phase-sensitive near-field microscope. The near-field probe, indicated by the dashed box, is scanned 20 nm above the sample and collects the evanescent field of the light inside the waveguide. The light is mixed with light from a reference branch. The resulting light is split by a polarizing beamsplitter, and the two orthogonal polarizing components are detected with a heterodyne scheme. By suitably choosing the orientation of the two \( \lambda/2 \) waveplates, we can relate the signal at the two detectors, called Ch1 and Ch2, with the fields present in the sample. (B) A scanning electron micrograph of two aluminum-coated near-field probes. For both probes, the coating thickness is 150 nm, and the aperture diameter is 200 to 230 nm. (Top) The highly cylindrical standard probe. (Bottom) A split probe in which an air gap in the metal coating (arrow) has been created.
longer shifted in space with respect to each other, as indicated by the dashed lines in Fig. 2F.

In the above, we only considered the amplitude of the detected signals. To understand the working of the split-ring probe, we also have to analyze the phase difference $\Delta \phi$ between the signals in Ch1 and Ch2. As mentioned above, the magnetic-field component of light induces a current in the ring, which in turn induces a time-varying charge distribution across the gap. Considering that we are far below the resonance frequency of the split probe [on the basis of methods for split-ring resonators (5), we estimate a resonant wavelength of $\lambda_0 = 1300$ nm and a width $\Delta \lambda_0 = 50$ nm for the resonance], the current in the ring should be in phase with the driving magnetic field. The resulting electric dipole moment along $\hat{x}$ will oscillate $90^\circ$ out of phase with the current and thus $90^\circ$ out of phase with the driving magnetic field (SOM text). In short, the probe will respond to $B_z$ with an electric dipole moment $p_x \propto iB_z$, which is analogous to the magnetic response of split-ring resonators (27, 11). To find the expected phase difference between the signals corresponding to the electric and magnetic field ($E_y$ and $B$, respectively), we use the curl equation $\nabla \times E = -\partial B/\partial t$. We know that for a plane wave traveling in the positive $x$ direction, $\omega B_z = -kE_y$, where $\omega$ is the optical frequency and $k$ is the wave number of the light. Thus, $B_z$ has a $\pi$ phase difference with respect to $E_y$. Given the additional $\pi/2$ phase shift induced by the probe, the signal detected in Ch2 should be $\pi/2$ out of phase with the $E_y$ signal detected in Ch1. Figure 3A shows for different probes and setup conditions (such as the strain on the fiber and the orientation of the connectors) that we indeed consistently measure a constant phase difference $\Delta \phi = -\pi/2$.

Fig. 2. (A), (C), and (E) Schematic of the performed experiments. In the upper part, top views are shown. The ‘ridge’ has been colored differently for clarity. In the lower part, cross-sections in the plane perpendicular to the substrate are shown. The red and green lines correspond to Ch1 and Ch2, respectively. (A) The standard probe is depicted in gray as a metallic ring. (C) The split probe is shown as a metallic split ring. (E) Same configuration as (C), but the waveguide is rotated by $90^\circ$. (B), (D), and (F) Line traces of the amplitude obtained by scanning a standard probe in the configuration shown in (A), a split probe in the configuration shown in (C), and a split probe in the configuration shown in (E), respectively, along the waveguide. (B) Both of the line traces, which are normalized to the maximum of Ch1, show a standing-wave component. (D) The line traces are normalized to the maximum of Ch2. The Ch2-detected signal is comparable with Ch1. We associate the Ch2-detected signal with $B_z$. (F) The line traces are normalized to the maximum of Ch2. Both $E_y$ and $B_z$ are projected along $\hat{x}$ and thus are detected by Ch2.

Fig. 3. (A) Phase difference $\Delta \phi$ between the complex signals of Ch1 and Ch2 for the measurement of Fig. 3B for different experimental conditions. (B) and (C) Normalized distributions of Ch1 $\propto \text{Re}(E_y)$ and Ch2 $\propto \text{Re}(iB_z)$, respectively. The images have been obtained by raster-scanning the split probe in the configuration shown in Fig. 2C over an area 2.2 by 3.4 m$^2$. The white dashed lines represent the position of the waveguide. The green dashed line is a guide to the eye that indicates the $\pi/2$ phase shift of the two wave fronts.
Characterization of a Rhodium(I) σ-Methane Complex in Solution

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Numerous transition metal–mediated reactions, including hydrogenations, hydrosilations, and alkane functionalizations, result in the cleavage of strong σ bonds. Key intermediates in these reactions often involve coordination of the σ bond of dihydrogen, silanes (Si-H), or alkanes (C-H) to the metal center without full scission of the bond. These σ complexes have been characterized to varying degrees in solid state and solution. However, a σ complex of the simplest hydrocarbon, methane, has eluded full solution characterization. Here, we report nuclear magnetic resonance spectra of a rhodium(I) σ-methane complex obtained by protonation of a rhodium-methyl precursor in CDC13 solvent at −110°C. The σ-methane complex is shown to be more stable than the corresponding rhodium(III) methyl hydride complex. Even at −110°C, methane rapidly tumbles in the coordination sphere of rhodium, exchanging free and bound hydrogens. Kinetic studies reveal a half-life of about 83 minutes at −87°C for dissociation of methane (free energy of activation is 14.5 kilocalories per mole).

Relative to \( \eta^2\)-H2 complexes, the synthesis and characterization of coordination compounds with saturated hydrocarbons acting as ligands, so-called σ-alkane complexes, have been particularly challenging (2–4). Not only is the strong nonpolar C-H σ bond a weak donor, but steric repulsions between the alkyl group and the metal center impede the close approach of the ligand to the metal center (5). Closely related to this interaction between an alkane and a metal are the now well-established intramolecular three-center, two-electron M\(^{+}\)-H2-C bonds of alkyl agostic complexes (6, 7). In these models for σ-alkane complexes, a C-H bond of an ancillary ligand is ideally positioned to interact with a metal center in a chelate-type interaction as shown in Fig. 1.

The existence of transition metal σ-alkane complexes as transient intermediates has been inferred by isotope scrambling studies and inverse kinetic isotope effects in the reductive

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References and Notes
18. Materials and methods and supporting text are available as supporting material on Science Online.
19. On one occasion, patterns have been visualized in a relatively complex nanostructure that resemble the calculated patterns of the magnitude squared of the magnetic field (30).