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Ferrimagnetic resonance excitation by light-wave mixing in a scanning tunneling microscope

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Ferrimagnetic resonance is measured in a scanning tunneling microscope. The infrared light of two lasers is focused into the tunneling junction and a difference-frequency signal in the microwave region is generated. This microwave signal is used to excite spin waves in an yttrium–iron–garnet film with a thin Au capping. The coupling of the light to the tunneling junction is explained by an antenna mechanism. Characteristic antenna patterns of the angle-dependent receiving efficiency are obtained. The mixing of the two laser frequencies is due to the nonlinearity of the tunneling junction. The microwave signal obtained is absorbed in the ferromagnetic sample if the resonance condition is fulfilled. This method might allow the measurement of magnetic properties with a lateral resolution down to the nm scale. © *1999 American Institute of Physics.*
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I. INTRODUCTION

The coupling of electromagnetic radiation into the tunneling junction of the scanning tunneling microscope (STM) offers the possibility to combine spectroscopic methods with the high lateral resolution and surface sensitivity of tunneling microscopy. This may lead to the identification and localization of adsorbed species at surfaces. First experiments demonstrating the spectroscopic capabilities of the STM have been done using microwaves[1] as well as radiation in the visible spectral range.[2] In this article we describe an application of this method to the detection of ferrimagnetic resonances in thin magnetic films using the STM.

In a ferrimagnetic resonance (FMR) experiment a microwave signal excites the precession of the spins in a magnetized sample, i.e., a spin wave with vanishing or small wave vector. From the determination of the resonance frequency as a function of an external magnetic field, important properties of the magnetism of the sample can be derived, e.g., the anisotropy or the gyromagnetic ratio.[3] In conventional FMR experiments the sample to be investigated usually forms an end wall of a microwave cavity. The probed area is then of the order of 1 cm^2. In recent experiments the sensitivity has been increased by using a microwave transmission line device that allowed the investigation of samples with an area of the order of several $\mu\mathrm{m}^2$.[4] The possibility of detecting electron spin resonance of paramagnetic spin centers using a STM has been demonstrated.[5,6]

In our experiment two infrared laser beams of slightly different wavelengths are focused into the STM tip.[7] The microwaves necessary for the excitation of the spin precession are produced by difference-frequency generation directly in the STM. The tunneling junction itself thus acts as a highly localized source of microwave radiation. In the case of a resonance in the sample, part of the microwave radiation is absorbed. It has been shown before that pictures taken with this microwave signal have atomic resolution.[8]

We demonstrate the excitation of spin waves in an yttrium–iron–garnet (YIG) sample using this difference-frequency signal. In addition, we investigate the coupling of the laser radiation into the tunneling junction of the STM as a function of the incidence angle in order to optimize the microwave signal.

II. EXPERIMENT

The experimental setup is shown schematically in Fig. 1. It consists of a STM, a laser system with two CO_2 lasers, a microwave detection system, and an electromagnet for applying an external magnetic field parallel to the sample plane.

The CO_2 lasers are operated at a wavelength close to 9.3 μm in the infrared. Gas fillings with different isotopes ($^{12}C^{16}O_2$ and $^{12}C^{18}O_2$) allow the tuning of the lasers to different transitions of the CO_2 molecule in order to obtain difference frequencies of several GHz. We performed measurements with difference frequencies of 2.15, 5.06, 9.03, and 13.2 GHz. Others are accessible with other combinations of isotopes. With pairs of polarizers the output power of each laser has been adjusted to 80 mW. The two laser beams were superimposed at a beam splitter. One of the two resulting beams is directed onto a photodetector in order to obtain a reference signal for the microwave detection system. The other beam is focused with a ZnSe lens with a focal length of 95 mm into the tunneling region of the STM. The focus has

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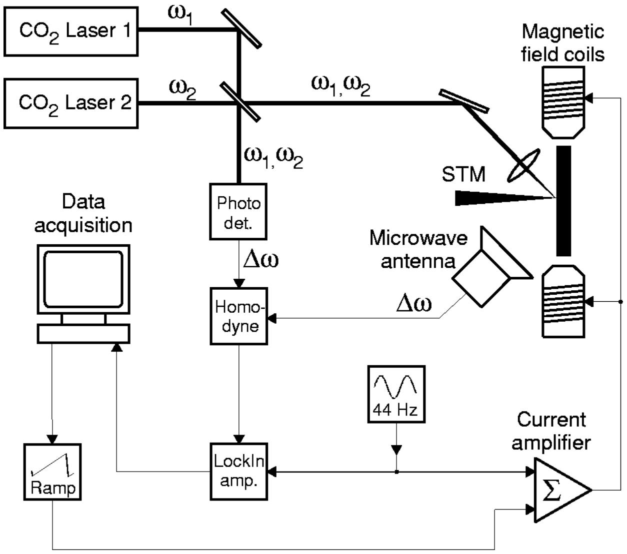

FIG. 1. Experimental setup.

a diameter of about 100 μm. The focusing lens is mounted on an *xyz*-translation stage in order to adjust the focus with the help of a red tracer beam from a HeNe laser. The polarization of the infrared light is parallel to the plane defined by the tip and the incident beam.

The tips used for the STM are etched from 0.2-mm-diam. tungsten wire by an electrochemical process. A scanning electron microscope image of a tip is shown in Fig. 2(b). The STM itself was built without any magnetic parts and even as few metallic parts as possible in order not to be sensitive to the modulated external magnetic field. The samples investigated were highly oriented pyrolytic graphite (HOPG) and a thin film (16.6 μm) of YIG on a gadolinium–gallium–garnet substrate. A 50 nm gold layer on the YIG sample provides electric conductivity for the operation of the STM.

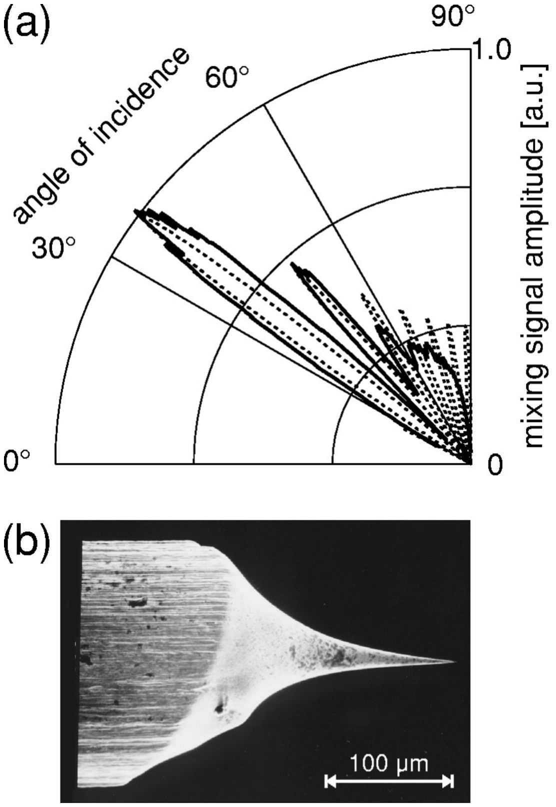

FIG. 2. (a) Experimental antenna pattern (solid line) for the STM tip shown in (b). The dashed line is a theoretical pattern for an ideal long-wire antenna.

A small microwave signal at the difference frequency of the two laser frequencies is radiated from the tip. For the detection of this signal we use a microwave horn antenna followed by a homodyne detection system. The setup of this system has been described in detail elsewhere.[9] Only minor changes to the initial setup have been made: for convenience of the adjustment, the metal–insulator–metal (MIM) diode for the generation of the local-oscillator signal has been replaced by a Hg–Cd–Zn–Te photodetector. Additionally, the appropriate microwave preamplifiers and mixers had to be chosen, depending on the frequency to be detected. With the homodyne technique we are able to measure signals as small as 10^{-18} W.

For the measurement of the dependence of the difference-frequency signal on the angle between the tip axis and the incident laser beam, the STM has been placed on a motorized rotation stage with an additional *xy*-translation stage. The rotation axis is perpendicular to the plane defined by the tip and the laser beam. With the translation stage the tunneling junction has been adjusted to lie in the rotation axis within less than 5 μm, as has been checked with a light microscope with a reticle. Thus, the displacement of the junction during rotation is small compared to the focus diameter of the infrared light.

The change of the amplitude of the microwave signal produced by the variable external magnetic field is measured with a lock-in technique. An electromagnet capable of generating fields up to 6.5 kOe is driven by a bipolar operational amplifier in the current mode. A voltage ramp is applied to the amplifier input in order to slowly increase the current through the magnet. A modulation voltage at 44 Hz is superimposed on the ramp. This voltage serves as the reference signal for the lock-in detection. All experiments have been performed under ambient pressure and temperature conditions.

III. RESULTS

The first experiment has been set up in order to investigate the coupling mechanism of the light to the tunneling junction. For this purpose the STM including the microwave horn were rotated continuously as described in Sec. II while the focus of the laser beam remained fixed on the tunneling region. The angle between the tip and the incident beam was varied between 20° and 90°. Simultaneously the difference-frequency signal was measured. The sample was HOPG. The result is shown in a polar plot in Fig. 2(a) together with the tip used for this measurement [Fig. 2(b)].

The FMR experiments were performed with the gold-coated YIG sample. The lasers were tuned to provide a specific difference frequency, and the magnetic field of the external electromagnet was increased from zero to its maximum value. The excitation of a ferrimagnetic resonance

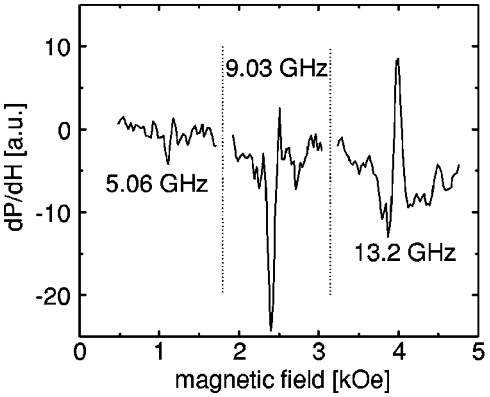

FIG. 3. Ferromagnetic resonance in the mixing signal. (P=amplitude of the difference-frequency signal; H=magnetic field).

leads to the absorption of the microwave signal at a specific value of the magnetic field. Using lock-in detection as described above, we measure the derivative of the microwave amplitude with respect to the magnetic field. Resonance curves for different mixing frequencies are shown in Fig. 3. The mixing-signal power received by the horn antenna was about 10^{-17} W. The time constants chosen for the measurements were 1 ms for the homodyne detection and 1 s for the lock-in amplifier. The slow field increase was repeated five times for each frequency. Figure 3 shows the averaged signals. The microwave absorption at the resonance at 13.2 GHz deduced from Fig. 3 is of the order of 1% of the mixing signal, at 5.06 GHz it is about four times smaller. At 2.15 GHz no resonance could be detected. The resonances at 5.06, 9.03, and 13.2 GHz are plotted as solid squares in Fig. 4 as a function of the magnetic field. Additionally, in Fig. 4 we show resonances obtained from a reference experiment where a microwave signal was directly coupled to the sample from a microwave synthesizer, and the absorption was measured (open squares).

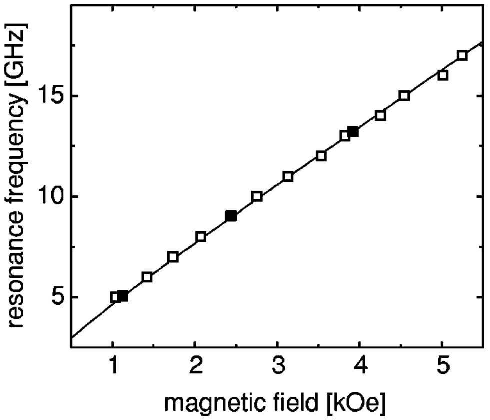

FIG. 4. Resonance frequencies obtained from the STM measurements (solid squares) and from the reference measurement (open squares) as a function of the magnetic field. The line is a theoretical curve as obtained with the material parameters of YIG.

IV. DISCUSSION

The difference-frequency generation in the STM is explained in three steps: The first is the coupling of the light into the tunneling junction of the STM. Then, nonlinear current components are generated, in our particular case, also a component at the difference frequency. In the last step the difference-frequency signal is radiated by the tip. In the case of a resonance in the sample, a part of the microwave radiation is absorbed.

The coupling of the laser light to the tunneling junction has first been described for MIM diodes.[10] Antenna patterns have been measured for laser wavelengths of 337 μm (Ref. 10) and 10.6 μm.[11] Compared to these wavelengths. the end of the tip is a thin wire and can be treated as a long-wire antenna for the light wave. Similar to the MIM diodes, in the case of the STM we have to consider an antenna in front of a reflecting plane (the sample) which acts as a mirror and thus doubles the antenna length. One obtains a radially symmetric antenna pattern with lobes in the polar direction. The angular dependence of the power received or emitted by the antenna is given by[12]

$$G(\theta)=\left(\frac{\cos(kL)-\cos(kL\cos\theta)}{\sin\theta}\right)^2. \tag{1}$$

Here, L is the effective antenna length, θ is the angle between the antenna axis and the incident beam, and $k=2\pi/\lambda$. The number of lobes in the half space is determined by L in terms of the wavelength λ.[11] The mixing signal amplitude is proportional to $G(\theta)$.[9]

A theoretical radiation pattern of an ideal long-wire antenna obtained from Eq. (1) is shown superimposed on the experimental curve in Fig. 2. The antenna length of the theoretical fit was chosen so that the lobe positions of the experimental data were matched. The result for the effective antenna length is $L=8\lambda$, corresponding to 74 μm. The broadening of the lobes as compared to the ideal pattern is ascribed to the conical form of the tip and the cone angle of the focused laser beam of 5.4°. Note that we omitted the main lobe of the pattern at about 20° since it is obscured by parts of the experimental setup and by the unetched part of the tungsten wire.

Such antenna patterns have been obtained for several tips. They all showed the lobe structure characteristic for long-wire antennas. Tips which were slightly bent in the first few micrometers still showed the lobes. Their mixing signal, however, was reduced by a factor of about 10. Obviously, the bending influenced the signal coupling to the tunneling junction significantly. Therefore, the tips used for the experiments were carefully selected using electron micrographs. Owing to the lobe structure, the correct angular position of the incoming light had to be adjusted in order to obtain large mixing signals. As can be seen from Fig. 2, the difference between the mixing signal at the first lobe and at the adjacent minimum is larger than a factor of 10.

The generation of new current components in the STM oscillating at the laser difference frequency is due to the nonlinearity of the current–voltage characteristics of the tunneling junction.[9,13] We use these difference-frequency signals as a localized source of microwave radiation in order to investigate magnetic properties of low-dimensional magnetic systems.

The microwave field excites the precession of the spins which leads to an absorption of radiation. The response of a magnetic moment $\boldsymbol{\mu}$ on a field can be described by the torque equation

$$\frac{1}{\gamma}\frac{d\boldsymbol{\mu}}{dt} = -(\boldsymbol{\mu}\times\mathbf{H}_{\mathrm{eff}}), \tag{2}$$

where γ is the gyromagnetic ratio and $\mathbf{H}_{\mathrm{eff}}$ is the effective magnetic field including the external field, the anisotropy field, and the time-dependent microwave field.

For our geometry with the magnetic field applied parallel to the sample plane, the resonance condition for uniform spin waves with small wave vectors is given by (see, e.g., Ref. 3)

$$\left(\frac{\omega}{\gamma}\right)^2 = H(H+4\pi M_s), \tag{3}$$

where $\omega/2\pi$ is the resonance frequency, H the external magnetic field, and M_s the saturation magnetization.

The parameters for a thin YIG film are $\gamma/2\pi = 2.8$ MHz/Oe and $4\pi M_s = 1750$ G.[14] The crystalline anisotropy field has been neglected since it is small compared to the external fields applied in the experiment.[15,16] The line in Fig. 4 shows the theoretical resonance condition given by Eq. (3). The experimentally observed resonances are exactly at the expected values.

V. CONCLUSION

Our experiments introduce a method to measure FMR by means of a STM using microwave signals directly generated in the tunneling junction by laser frequency mixing. The coupling of the laser light to the tunneling junction has been understood by an antenna model that leads to a characteristic antenna pattern. The experiments reported here are laser-assisted STM measurements of sharp resonances characteristic for the material under investigation. In principle, the method can be extended to other wavelength ranges by using also the incident laser frequency for resonant excitation. In this way vibrational or electronic transitions become accessible.

The technique has been tested on a continuous YIG film. It is, however, highly desirable to use this method for the microscopic imaging of the magnetic properties of small magnetic structures as, e.g., magnetic nanowires.[17] For measurements of the dynamical response of a spin system, the highest resolution is currently achieved with Brillouin light scattering where diffraction of light sets the limit to about 0.5 μm. The long measurement times necessary for the resonance detection in the present experimental configuration preclude the generation of images in order to show experimentally the high lateral resolution that is expected from this method. The necessary increase of the detection efficiency could be achieved by integrating the tunneling junction of the STM into a microwave cavity.[18] An estimate shows that in such an experimental setup[9,19] the detected microwave signal generated by the same oscillating tunneling current can be increased by several orders of magnitude.

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