Laser-Driven Scanning Tunneling Microscope

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New modes of operation of a scanning tunneling microscope (STM) with laser radiation coupled into the tip of the tunneling junction are demonstrated. The dc current generated by rectifying the laser light and the difference-frequency signal produced in the STM by two laser beams are used to obtain atomic-resolution surface images of graphite, as well as to control the tip-sample distance. Such a laser-driven STM can generate surface images without external bias voltage and, when the difference-frequency signal is used for the distance control, even without any dc current between the tip and the sample. This mode of operation may also allow insulators to be studied.

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The scanning tunneling microscope (STM) was the starting point for the development of numerous new methods to obtain atom-resolved surface information. In the usual mode the STM produces images of the local density of states at the surface.[1] New modifications of the STM allow the generation of surface images with quantities such as atomic forces,[2] photon emission,[3] temperature,[4] and ion conductance,[5] as well as images with spectroscopic information.[6] In some of these experiments new modes of operation have been introduced where the distance dependence of these quantities is used to control the width of the tunneling gap of the STM.[2,4,5]

In this Letter we report on experiments where laser radiation is coupled into the tip of the STM. Owing to the nonlinearity of the current-voltage (I-V) characteristic of the tunneling junction laser, light rectification and difference-frequency generation of two injected laser signals are observed. The nonlinearity of the I-V curve depends on the atomic structure of the sample. As a consequence, when a surface scan is performed, both the rectification signal and the difference-frequency signal reproduce the atomic surface corrugation. In addition, the gap-width dependence of these signals offers the possibility to control the tunneling distance by using either one of these signals. In this way a laser-driven tunneling microscope can be realized. Using CO_2 laser radiation, we successfully demonstrated both new schemes for distance control and obtained atomic-resolution images of a graphite surface by recording either the rectified or the difference-frequency signal.

The interaction of laser radiation with the tunneling junction of the STM can be regarded as analogous to the interaction with the metal-insulator-metal (MIM) point-contact diode.[7] As with the MIM diode, the laser radiation is coupled into the tunneling junction of the STM via the fine metal tip which, for infrared wavelengths, acts as a long-wire antenna. This leads to a voltage between the tip and the surface oscillating at the laser frequency, and, in addition, to heating and thermal expansion of the tip. Because of the nonlinear current-voltage characteristic of the tunneling junction, new current components arise, e.g., at zero frequency (rectification), and if two laser beams with different frequencies are injected frequency mixing is also observed. These laser-induced current components are proportional to the second derivative of the I-V characteristic $\partial^2 I/\partial V^2$.

In previous experiments we demonstrated difference-frequency generation in the STM up to the THz range[8] by using CO_2 laser radiation with wavelengths close to 10 μm, and we observed rectification of the laser radiation.[9] It was shown that the generation of the difference-frequency signals at 9 GHz is determined by the nonlinearity of the static I-V curve of the tunneling junction.[9] Furthermore, we confirmed that these nonlinear components are dependent on the width of the tunneling gap. Decreasing the tip-sample distance leads to an increase of the mixing signal. The starting point for the present experiments was the additional observation that the mixing-signal intensity as well as the rectified laser signal reproduce the atomic structure when the tip was scanned across a graphite surface.[10]

The experimental setup for the generation of laser difference frequencies in the GHz range and the measurement of laser light rectification has been described in detail elsewhere.[10] The main components of the experiment are the STM, two single-mode line-tunable CO_2 lasers, the microwave detection system for the mixing signal, and a computer for data acquisition. A schematic drawing is shown in Fig. 1(a).

For the tunneling junction a highly oriented pyrolytic graphite surface and tungsten tips are used. The experiments are carried out at ambient air pressure. A frequency difference of 9 GHz is provided by mixing two laser beams with a wavelength close to 9.3 μm produced in two lasers operated with CO_2 of different isotopic compositions ($^{12}C^{16}O_2$ and $^{12}C^{18}O_2$). The two beams

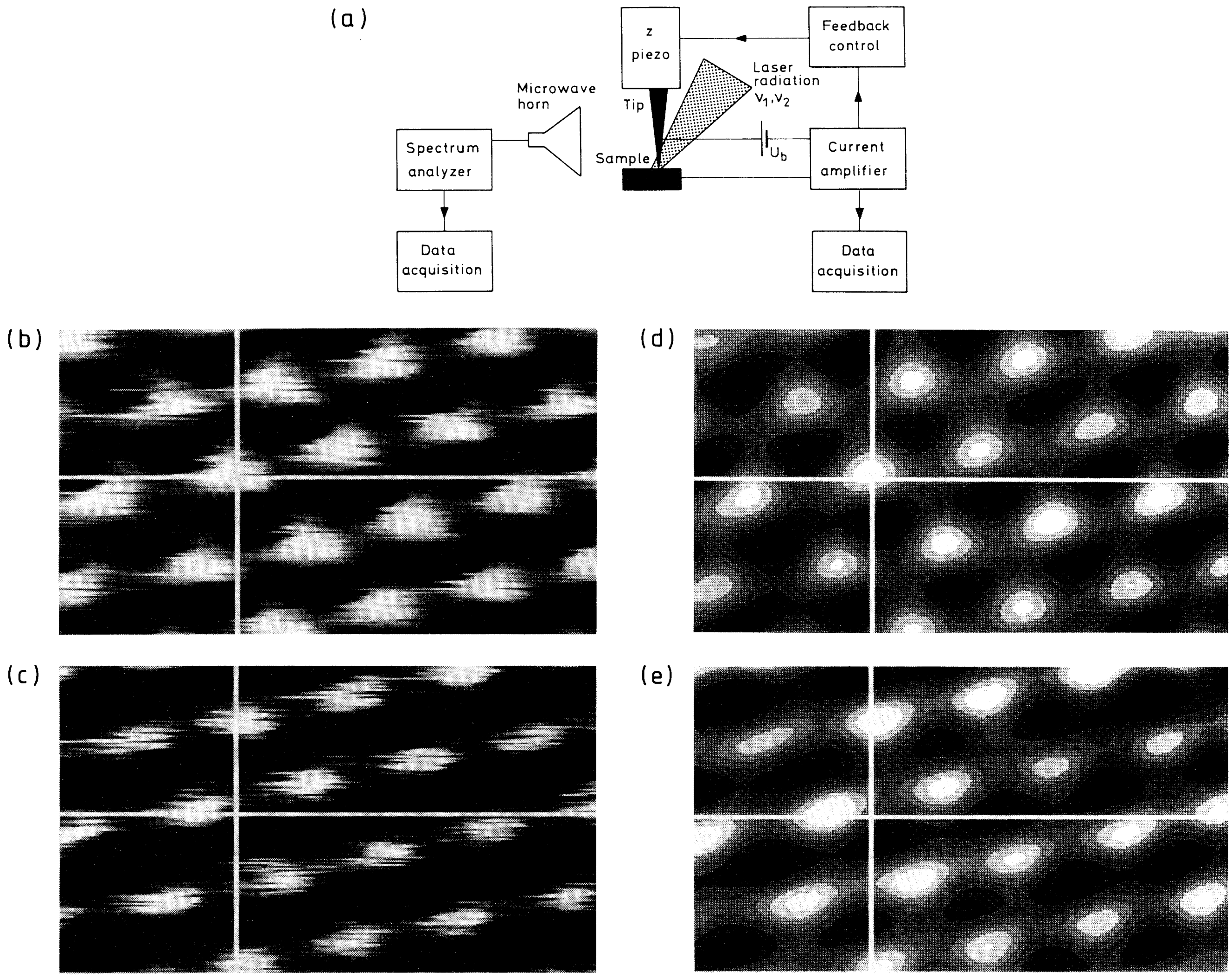

FIG. 1. (a) Schematic drawing of the experimental setup for the generation and detection of laser difference frequencies $\Delta\nu = \nu_1 - \nu_2$ in the tunneling junction of the STM (U_b represents bias voltage). Images of a graphite surface obtained with this setup by simultaneously recording (b) the tunneling current and (c) the difference-frequency signal. The parameters are preset current 50 nA, tip bias voltage −40 mV, total laser power 30 mW, and $\Delta\nu = 9$ GHz. (d),(e) The raw data of (b) and (c), respectively, after filtering in Fourier space.

are focused collinearly onto the tip at an angle of about 30° to the tip axis. A total laser power of 30 mW is used with a spot diameter at the tip of about 60 μm.

The mixing signal (typically 10^{-12}–10^{-15} W) is collected with a microwave horn, amplified, and detected with a spectrum analyzer. The dc current due to rectification of the laser radiation is measured at zero bias voltage with one laser beam focused into the tunneling junction. In order to keep the tunneling distance constant at zero bias voltage the feedback loop is opened for each measurement. The current amplifier normally used to measure the tunneling current was also used to measure the rectified current. The value of this current can reach the same order of magnitude as the preset tunneling current. As an example, at 30 mW of incident laser power and a gap width determined by a preset current of 5 nA and a tip bias voltage of +30 mV the current due to rectification at zero bias voltage is about 1.5 nA with the same sign as the preset current.

When a surface scan is performed, surface images can be obtained by recording either the tunneling current, the rectification signal, or the mixing signal. Figure 1(a) shows a schematic drawing of the experimental setup for a simultaneous recording of graphite surface images using the tunneling current and the mixing signal intensity at 9 GHz. The corresponding images are displayed in Figs. 1(b) and 1(c), respectively. For these images the tip-sample distance is controlled by keeping the average tunneling current at 50 nA with a tip-bias voltage of −40 mV. For these tunneling parameters the maxima (bright spots) of the tunneling current and of the mixing signal occur at different locations on the graphite surface, as can be recognized by means of the two intersecting lines. Similar images are obtained by recording the tunneling current and the current due to rectification.[10]

In a second step the rectified signal is used to control the tip-sample distance in the STM. To set up this mode of operation the first tip-sample approach is, as usual, controlled by the tunneling current. Next, the rectified current is measured as described above, and the focus adjustment on the tunneling junction is optimized by maximizing this current. Finally, the rectified current is used as input signal for the feedback control of the distance in the tunneling junction, as shown in Fig. 2(a). Since in this setup the laser generates a current in the STM, no externally applied bias voltage is necessary. An image of the graphite surface obtained in this mode of operation by monitoring the rectified signal is shown in Fig. 2(b). For this image the average tip-sample distance is determined by a rectified current of 1 nA generated by a laser power of 30 mW. High values of the current correspond to the bright spots in the figure.

In a further experiment we used the intensity of the 9-GHz difference-frequency signal to control the tip-sample distance and to generate surface images. In this case the STM is not only receiving but also emitting electromagnetic radiation. As we use the emitted radiation for distance control as well as for imaging, no external bias voltage and no current measurement are necessary. All electrical connections to the sample can therefore be removed. A schematic drawing of the experimental setup is presented in Fig. 3(a). In this circuit the spectrum analyzer serves as a narrow-band detector at a fixed frequency and its output is connected to the feedback control. The distance is adjusted so that the average signal is kept at a preset level. With this configuration images of the graphite surface are obtained by recording the intensity of the mixing signal. An example is displayed in Fig. 3(b).

As an essential feature the surface images generated by the laser-induced current components demonstrate atomic resolution. This is expected since in addition to the tunneling current the second derivative $\partial^2 I/\partial V^2$ also varies on an atomic scale.

The location of the maxima in the laser-generated images is determined by the properties of the I-V characteristic and their dependence on the bias voltage and the tip-sample distance (scanning tunneling spectroscopy).

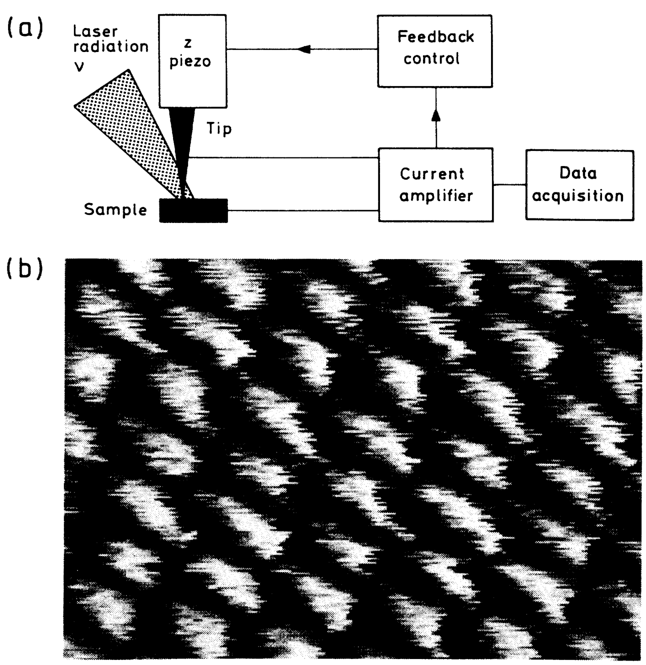

FIG. 2. (a) Schematic drawing of a laser-driven STM whose tip-sample distance is controlled by the rectified current. Note the absence of a bias voltage source in the tunneling circuit. (b) Image of a graphite surface obtained with this setup by recording the rectified current. The parameters are preset rectified current 1 nA and laser power 30 mW. No filtering is applied.

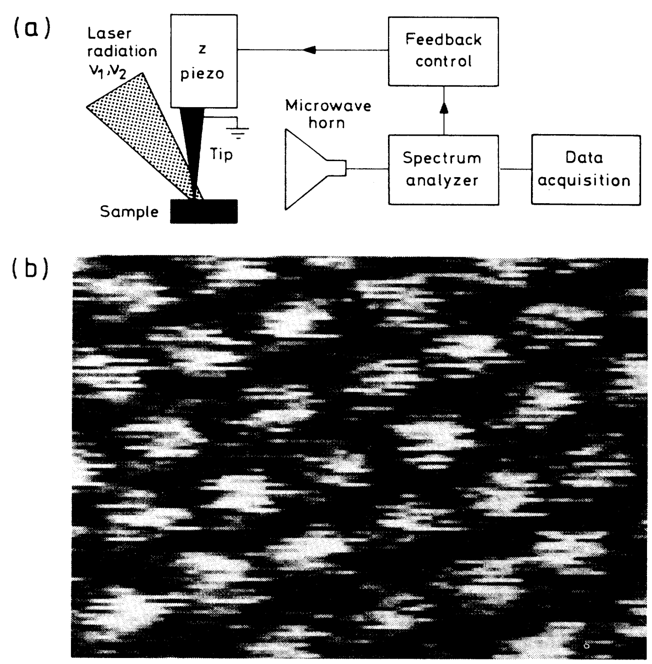

FIG. 3. (a) Schematic drawing of a laser-driven STM whose tip-sample distance is controlled by the difference-frequency signal of two injected laser signals at ν_1 and ν_2. Note the absence of any electrical connection to the sample. (b) Image of graphite surface obtained with this setup by recording the difference-frequency signal at $\Delta\nu = 9$ GHz with a total laser power of 30 mW. No filtering is applied.

In the images of Fig. 1 the maxima of the tunneling current and of the difference-frequency signal are shifted with respect to each other. They may, however, also coincide. We have observed different relative positions at different values of the bias voltage.[10]

The atomic resolution of the newly generated surface images also points to the fact that the tunneling junction of the STM is a nonlinear device of microscopic dimensions and therefore has an extremely small response time. This is demonstrated by the generation of difference frequencies in the THz range[8] and by the rectification of CO_2 laser radiation at 32 THz.

When laser radiation is coupled into the tip of the tunneling junction, the tunneling distance in the usual current-controlled STM may be crucially changed by additionally generated current components. Besides the rectified current, thermocurrents are also produced, because the tunneling junction acts as a thermocouple when tip and sample consist of different materials; the higher temperature of the tip leads to a thermoelectric current even when the two materials are identical. In our experiment the thermoelectric currents are expected to flow from the sample to the tip. The observed laser-induced dc current, however, flows in the opposite direction. The thermoelectric currents are therefore overcompensated by a dominating current contribution which is due to laser light rectification.

These laser-induced dc current components add to the normal bias-induced tunneling current. In the laser-driven STM the number of distance-dependent current components is reduced since the microscope is operated without a bias-induced tunneling current. No dc current component at all is necessary when the intensity of the difference-frequency signal controls the tunneling distance. A tunneling microscope also operating without dc current was recently realized in a different way by Kochanski.[11] In this experiment considerably lower frequencies in the microwave region were used.

If laser difference-frequency generation is also observed on nonconducting surfaces, the laser-driven STM is expected to generate atomic-resolution images of insulators. This would extend the applicability of the STM to a whole new class of materials.

In the present experiment the bias-induced tunneling current is still used for the tip-sample approach and for the distance control at the beginning when the nonlinear current components are optimized. For a systematic investigation of difference-frequency generation on nonconducting surfaces a force measurement as used in the atomic-force microscope could be applied for the initial tip-sample control. These experiments are in progress.

The surface images generated by the nonlinear current components can be used in conventional scanning tunneling spectroscopy since the second derivative of the current is very sensitive to small current changes. The most important application of the laser-driven STM, however, lies in optical spectroscopy at surfaces. The laser radiation introduces a new parameter into tunneling microscopy: the photon energy. Measuring with different laser frequencies allows us to investigate adsorbate-specific resonances or excitations in the surface. These resonances can be excited and localized with the laser-driven STM with atomic resolution.

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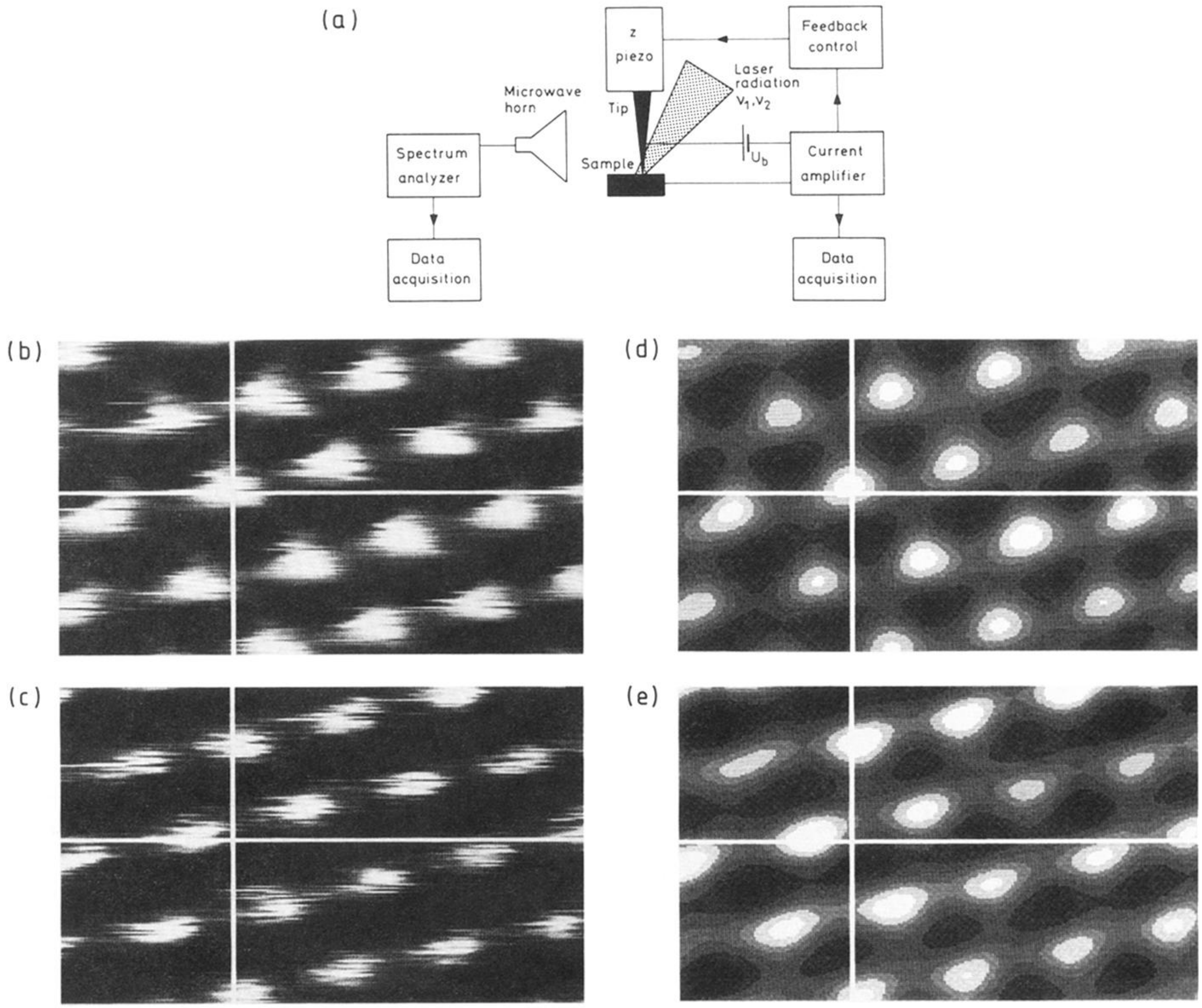

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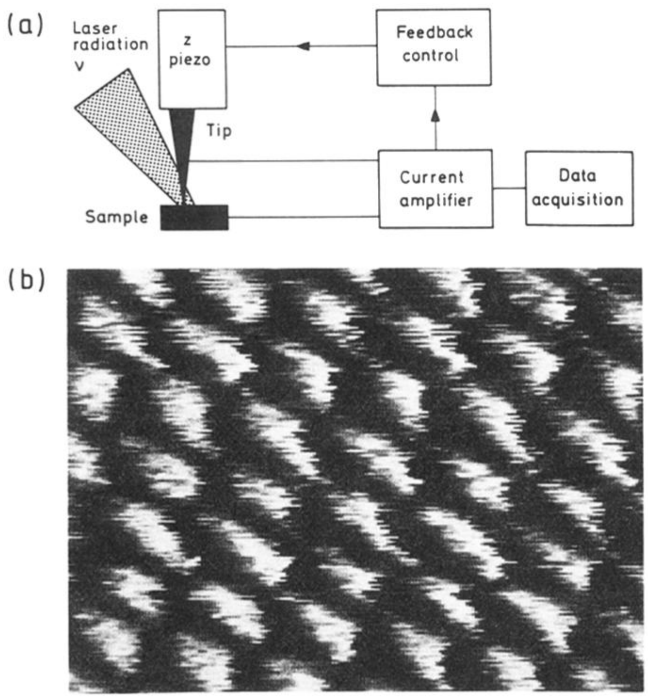

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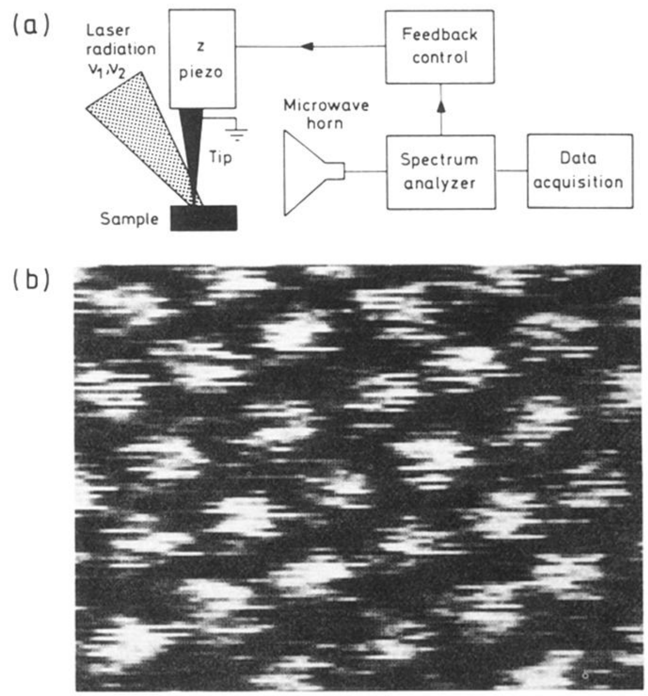

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