Doppler-Free Two-Photon Polarization-Spectroscopic Measurement of the Stark-Broadened Profile of the Hydrogen L_α Line in a Dense Plasma

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It is demonstrated that Doppler-free two-photon polarization spectroscopy makes possible Stark profile measurements without Doppler-broadened background under high-density and high-temperature conditions, e.g., in dense arc plasmas, where other sub-Doppler laser-spectroscopic techniques introduced so far fail. This method, based on the polarization rotation connected with a two-photon transition, is not limited by collisions changing the velocity or the excitation of the atoms or by photoionization of the upper level.

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Broadening and splitting of spectral lines due to the Stark and Zeeman effect provide valuable tools for investigations in the field of plasma physics. In those high-temperature environments, however, the Doppler effect due to the thermal motion of the atoms is often the dominating cause of line broadening; thus the phenomena of interest are hidden by the overwhelming Doppler broadening.

On the other hand, Doppler-free spectroscopic techniques introduced so far with various detection schemes for different applications, e.g., saturation spectroscopy,[1] polarization spectroscopy,[2] or two-photon spectroscopy,[3,4] cannot be applied to dense plasmas (see, e.g., Weber, Frankenberger, and Schilling[5] and Goldsmith[6]) because of the comparatively high collision rates for inelastic and elastic collisions or because of the strong photon emission of the plasma. The signals to be detected for those sub-Doppler techniques would be destroyed by collisions changing the velocity or excitation of the atoms or would be hidden by the strong radiation background. To overcome these limitations we introduce a Doppler-free two-photon polarization-spectroscopic technique which makes plasmas accessible for Doppler-free spectroscopic investigations and diagnostics, for a considerably wider range of parameters. The method is demonstrated by a Doppler-free two-photon L_α Stark profile measurement in a dense arc plasma with an electron density of 5×10^{22} m^{-3} and a temperature of 11 000 K. For these plasma parameters a reliable test of the new technique is possible by comparison with Stark-profile calculations[7] which are well proved experimentally[8,9] for one-photon Stark profiles.

The overall scheme of Doppler-free two-photon polarization spectroscopy described in this paper looks rather similar to the familiar one-photon polarization spectroscopy.[2] A linearly polarized probe beam becomes slightly elliptically polarized by passage through a sample with an optical anisotropy induced by a circularly polarized pump beam. As a principal difference, though, in our case the probe beam experiences the induced susceptibility of a two-photon transition.[10] The polarization rotation to be detected is created at the instance of the two-photon transition; hence velocity-changing and excitation-changing collisions do not destroy the signal. Two-photon polarization spectroscopy can therefore be applied to high-density and high-temperature environments and, in fact, requires comparatively high densities because of the small two-photon transition probabilities.

The hydrogen L_α transition is well suited for a demonstration of the capability of the two-photon polarization spectroscopy because the selection rules for the $1S$-$2S$ transition only allow for the absorption of two photons of opposite circular polarizations. With a right-hand circularly polarized pump beam tuned to half the frequency of the L_α transition only the right-hand component[11] of the counterpropagating linearly polarized probe beam is influenced by the susceptibility induced by the pump beam. The resulting polarization rotation of the probe beam can be measured behind a crossed polarizer. Because only absorption of one photon from each beam provides the signal, the first-order Doppler shifts cancel and there is no Doppler-broadened background. The detection sensitivity is particularly limited by the residual transmittance still present for perfectly crossed polarizers and by the amount of depolarizing scattering occurring in the experimental setup.

The experimental arrangement is shown in Fig. 1. The second harmonic of a Q-switched Nd-doped yttrium-aluminum-garnet laser is used to pump a dye laser operating at 630 nm. The dye-laser second harmonic (315 nm) is mixed with the Nd-doped yttrium-aluminum-garnet fundamental (1064 nm) to produce tunable radiation at 243 nm of about 1 mJ per pulse with a pulse width of 5 ns at a repetition rate of 10 Hz. Approximately 10% of the 243-nm radiation is split off to serve as the linearly polarized probe beam. The remaining 90% is sent through a quartz quarter-wave plate to become the circularly polarized pump beam. The counterpropagating beams are focused by lenses

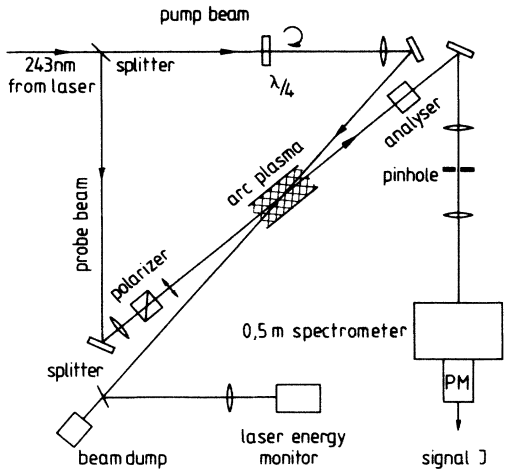

FIG. 1. Experimental setup.

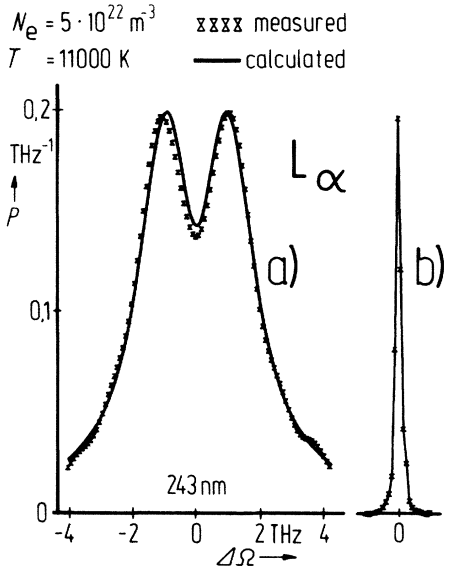

FIG. 2. (a) Measured and calculated two-photon L_α Stark profiles $P(\Delta\Omega)$ vs the angular frequency detuning of the laser off resonance. For the comparison the measured profile was area normalized with respect to the calculated profile. The measured profile was obtained as the average of four independent scans over 128 frequency points with a step size of 86 GHz angular frequency corresponding to 2.68 pm at 243 nm and 100 laser shots per point. (b) Two-photon L_α profile displaying the spectral resolution: FWHM is 160 GHz angular frequency or 5 pm at 243 nm. This profile was obtained from a plasma region with low electron densities where Stark broadening is negligible.

of 1000-mm focal length into the center of the arc plasma (8 mrad crossing angle). The probe beam is adjusted on the axis of the plasma column. The measured spot size at focus is 300 μm; this corresponds to an irradiance of about 300 MW cm^{-2}. After passing through the crossed analyzer the probe beam is imaged into a 0.5-m grating spectrometer to eliminate the strong background emission of the arc plasma. A small part of the pump beam is split off for monitoring of the energy of each laser pulse. The plasma is created at atmospheric pressure in a wall-stabilizing cascaded arc[9] with a channel diameter of 4 mm at discharge currents of 30 A. A mixture of 50% argon and 50% hydrogen is fed into the 50-mm-long center section. The end sections and the electrodes are operated in pure argon. In order to avoid undesirable depolarization of the probe beam by windows closing the arc chamber, those are substituted for by an argon-gas flow from the arc chamber into the surrounding atmosphere. Thus for the experimental setup a residual transmittance of 3×10^{-7} was achieved for perfectly crossed air-spaced Glan polarizers. The experiment is controlled by a microcomputer which steps the dye laser, reads the signals, and performs the data evaluation as described in the following section.

For the two-photon L_α transition the absorption of the pump and the probe beam as well as the polarization rotation of the linearly polarized probe beam are small. With perfectly crossed polarizers the photomultiplier signal I measured behind the spectrometer is given therefore by the following expression[12]:

$$I - I_{\rm res} \sim [d|\chi^{(3)}(\Delta\Omega)|E_c]^2 E_l. \tag{1}$$

E_l is the irradiance of the linearly polarized probe beam in the probe volume of length d and E_c is the corresponding irradiance of the circularly polarized pump beam. $\chi^{(3)}(\Delta\Omega)$ is the two-photon L_α-resonant part of the third-order nonlinear susceptibility, strongly depending on the circular frequency detuning $\Delta\Omega$ off resonance. $I_{\rm res}$ is the residual part of the photomultiplier signal accounting for the residual transmittance and was measured with the probe beam kept on the axis of the arc-plasma column while the pump beam passed through the plasma without crossing the probe beam.

According to Ref. 12 the frequency dependence of $|\chi^{(3)}(\Delta\Omega)|^2$ can be expressed as

$$|\chi^{(3)}(\Delta\Omega)|^2 \sim N_1^2 |\tilde{C}(\Delta\Omega)|^2 \sim N_1^2 P(\Delta\Omega); \quad \int P(\Delta\Omega)\, d\Delta\Omega = 1. \tag{2}$$

N_1 is the number density of atomic hydrogen in the ground state and $P(\Delta\Omega)$ is the normalized two-photon L_α Stark profile which is proportional to $|\tilde{C}(\Delta\Omega)|^2$ provided by Stark-profile calculations.[12]

From the expressions (1) and (2) one obtains

$$P(\Delta\Omega) \sim (I - I_{\rm res})/E_l E_c^2, \tag{3}$$

which has been used in order to evaluate the two-photon L_α Stark profile $P(\Delta\Omega)$ from the measured

photomultiplier signals; i.e., for each laser pulse the difference $I - I_{res}$ was related to the third power of the irradiance provided by the laser which was determined from the monitored laser energy.

In Fig. 2(a) a Doppler-free L_α profile measured at an electron density $N_e = (5 \pm 0.5) \times 10^{22}$ m^{-3} and a temperature $T = 11\,000 \pm 500$ K is compared with a calculated two-photon L_α profile obtained by computer simulations.[12] The striking agreement between theory and experiment becomes evident, indicating that both the theoretical approach and the experimental technique are well understood. In contrast to the more familiar one-photon Stark-broadened L_α profile ($2P$-$1S$ transition), the Stark-broadened two-photon L_α profile has a dip in the center because only one red- and one blue-shifted Stark component contribute to the $1S$-$2S$ transition. The L_α profile was measured with the probe beam adjusted to a position on the axis of the arc-plasma column where the electron density varies by less than $\pm 2\%$ over the 300-μm laser spot size.

In Fig. 2(b) a much sharper Doppler-free L_α profile is shown to demonstrate the spectral resolution provided by the laser. This profile was obtained from low-electron-density plasma layers near the wall of the plasma chamber where Stark broadening is negligible compared to the laser linewidth. Therefore, the profile exhibits the spectral resolution which was found to be 5 pm FWHM at 243 nm or 160 GHz angular frequency. No shift between the high-density profile (a) and the low-density profile (b) could be observed within an uncertainty of ± 2 pm (± 64 GHz angular frequency).

Finally we summarize the advantages of two-photon polarization spectroscopy. In contrast to the usual one-photon polarization spectroscopy no change of atomic-level populations with subsequent probing is involved. The signal to be detected is created at the instance of the two-photon absorption; therefore collisions changing the velocity or the excitation of the atoms or photoionization of the upper level do not destroy the signal. The detection sensitivity is particularly limited by the residual transmittance still present for perfectly crossed polarizers and by the amount of depolarizing scattering due to the experimental setup. Because only absorption of one photon from each beam provides the signal, the first-order Doppler shifts cancel and there is no Doppler-broadened background. Two-photon polarization spectroscopy can be applied to atoms in high-density and high-temperature environments. This method, providing high spectral and spatial resolution, opens therefore a new field for spectroscopic investigations and diagnostics, e.g., of plasmas. Measurements of the L_α Stark broadening, for instance, become possible now for electron densities less than 10^{23} m^{-3}. These measurements are highly desirable, considering that new theoretical approaches[13,14] predict ion dynamical effects to increase the L_α Stark width at $N_e \approx 10^{21}$ m^{-3} by an order of magnitude.

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