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Measurements of saturation broadening of the Cu $5s'\,^4D_{5/2}$ autoionization level by the pulsed optogalvanic technique

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The use of the optogalvanic technique to obtain the line profile of the Cu $5s'\,^4D_{5/2}$ autoionization level with different degrees of saturation broadening is demonstrated. The experimental results point out that the growth of the autoionization-level breadth follows the square root of the increase in the laser intensity. The measured value of the natural breadth of the Cu$5s'\,^4D_{5/2}$ autoionization level is 7.2 ± 0.6 cm^{-1}. The laser saturation intensity was measured by two different methods, and its value is about 0.39 ± 0.09 MW/cm^2.

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I. INTRODUCTION

The optogalvanic effect is the change in the ionization rates and the dynamic conductivity of the plasma due to laser illumination tuned to excite one to the plasma ingredients. The optogalvanic effect is widely used in atomic and molecular spectroscopic techniques such as a Doppler-free spectroscopy [1], plasma diagnostics [2,3], fine and hyperfine structures in oxygen [4], high-resolution intermodulated optogalvanic saturation spectroscopy in heavy elements [5], Rydberg-state spectroscopy [6], diagnostics of metastable AR levels in rf glow discharge [7], and optogalvanic line profiles of several Kr atomic lines measured by Kawakita *et al.* [8]. The major restriction of using the optogalvanic technique in measuring atomic-line profiles is that the laser spectral width must be much narrower than the width of the detected state; therefore, Pfaff, Begemann, and Saycally [9] were unable to measure line profiles of Cu autoionization levels. Recently, we have been able to scan the resonance profiles of several Cu autoionization levels by using a Fabry-Pérot intercavity étalon inside a Hanch-type dye laser [10]. In the present Brief Report, we measured directly the saturation broadening of the Cu $5s'\,^4D_{5/2}$ autoionization level as a function of the dye-laser intensity.

II. EXPERIMENTAL SETUP

The experimental setup is displayed schematically in Fig. 1. It includes an excimer laser as a pumping laser (Lambda Physik EMG-201) with average power of 5 MW and pulse duration of 15 ns. It pumps a Hansch-type (Lambda Physik Fl-2000E) dye-laser consist of Coumarian 47 as an active dye solution. The dye laser emits visible light pulses at 458.7 nm having a pulse energy of a few millijoules and a linewidth of 0.15 cm^{-1} (with Fabry-Pérot intercavity étalon). The dye-laser illumination is aligned to the center of the hollow cathode zone of a Hamamatsu L-233 type Cu-Ne HCD (hollow cathode discharge) tube. The HCD tube is filled with about 5 torr of neon and operates at the steady state at a voltage below 500 V. The electrical circuit that activates the HCD tube includes a very stable power supply and a 20-kΩ load resistor to limit the tube current to less than 20 mA. In order to avoid rf noise, the HCD tube, the power supply, and the electrical circuit components are kept inside a Faraday cage. The optogalvanic signals were coupled via a dc blocking capacitor to a Hewlett-Packard 54020A digital oscilloscope and displayed on an ink jet printer. The dye-laser emission wavelength is monitored by a Spex 1401 double monochromator and a sensitive photodiode.

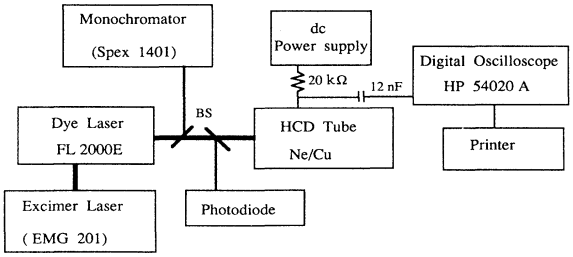

FIG. 1. Experimental setup used for measuring the optogalvanic line profiles.

III. THEORETICAL CONSIDERATIONS

The configuration $3d^04s(^3D)5s$ of the copper atom contains four levels: $5s'\,^4D_{1/2}$, $5s'\,^4D_{3/2}$, $5s'\,^4D_{5/2}$, $5s'\,^4D_{7/2}$. The interval-energy ratios [11] are 7:5:4. In a pure *LS*-coupling scheme, the interval-energy ratios would be 7:5:3. Therefore intermediate coupling is used to describe more adequately the interval-energy ratios of the highly excited levels of the copper atom. The copper autoionization levels $5s'\,^4D_{5/2}$ and $5s'\,^4D_{3/2}$ are 632 cm^{-1} and 1267 cm^{-1} above the ionization level of the Cu atom, which is 62 317 cm^{-1} above the ground state of Cu. The autoionization levels are very broad owing to configuration interaction, which couples the discrete atomic level to a band of continuum states belonging to a different configuration. The theoretical interpretation is due to Fano [12], who calculated the spectral distribution of the transition probability (or absorption strength) to a discrete state perturbed by a continuum.

The results can be expressed in terms of two parameters. The first is the value Γ of the width of the autoionization state, where $(\Gamma/\hbar)$ is the autoionization probability. The other parameter is q, which is the ratio of the

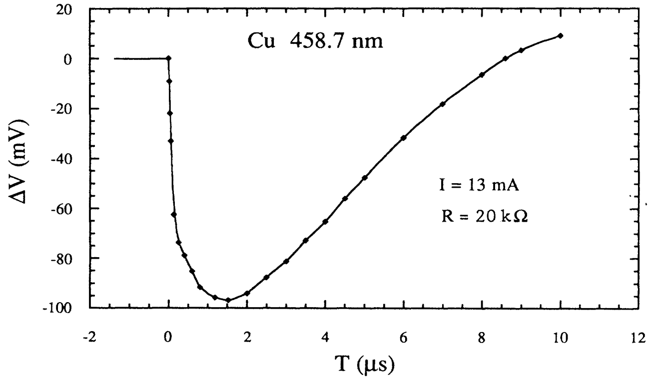

FIG. 2. Typical optogalvanic signal detected from the relaxation of the Cu $5s'\,^4D_{5/2}$ autoionization level.

matrix elements of the optical-transition operators to the perturbed discrete state and to the continuum state, respectively. Fano introduced a reduced scale for the energy difference relative to the position of the perturbed line by the following formula:

$$\varepsilon=2\hbar(\omega-\omega_0)/\Gamma+1/q\ , \tag{1}$$

where ω is the laser frequency and ω_0 is the autoionization resonance-transition frequency. According to Fano's theory, the autoionization line profile is given by the following equation:

$$\alpha(\varepsilon)=[q+\varepsilon]^2[1+\varepsilon^2]\ . \tag{2}$$

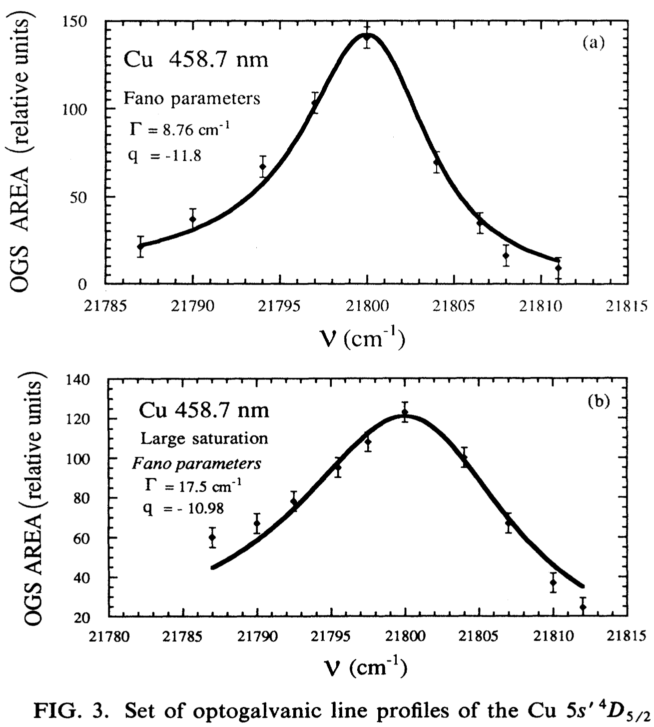

FIG. 3. Set of optogalvanic line profiles of the Cu $5s'\,^4D_{5/2}$ autoionization level measured at different degrees of saturation: (a) at moderate saturation, (b) at large saturation. The solid lines are the computer-fitted Fano-Beutler asymmetrical line profiles to the optogalvanic data points.

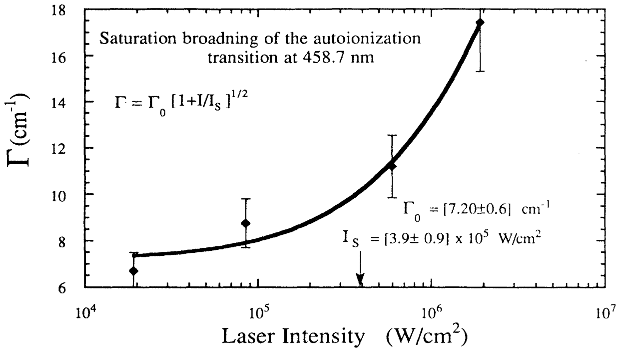

FIG. 4. Linewidth parameters as a function of the laser intensities used in exciting the Cu autoionization transition $4p'\,^4F^\circ_{7/2}\rightarrow 5s'\,^4D_{5/2}$ at 458.7 nm. The solid line is the fitted saturation relation $\Gamma=\Gamma_0(1+I/I_S)^{1/2}$, with the parameters $\Gamma_0=7.2\ \mathrm{cm}^{-1}$ and $I_S=3.9\times10^5\ \mathrm{W/cm}^2$.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

We have measured several optogalvanic line profiles of the Cu $5s'\,^4D_{5/2}$ autoionization level at variable dye-laser intensity. We have tuned the laser frequency around the resonance profile of the Cu $4p'\,^4F^\circ_{7/2}\rightarrow 5s'\,^4D_{5/2}$ transition at 1-cm^{-1} steps, and recorded the OGS (optogalvanic signal) generated from the ionization-relaxation channel of the Cu $5s'\,^4D_{5/2}$ autoionization level.

The detected optogalvanic signals possess a unique structure. It starts with a very fast voltage fall at its initial part, as presented in Fig. 2. This indicates that the population transferred to the autoionization level from lower levels by the laser radiation relaxes mainly through the ionization channel, and it occurs during the laser existence inside the discharge tube. Such kinds of signals have been presented by Shuker and Hakham Itzhaq [10]. Based on the assumption that the optogalvanic signals are proportional to the absorption coefficient of the optical transition, we have measured the temporal integrated area of each OGS. Substitution of these results as a function of the laser frequency enabled us to construct the optogalvanic line profile of this level, as shown in Fig. 3. We have used a special computer software to fit these data with the most approximate asymmetrical Fano-Beutler line profiles.

The computer fit the results in Fano parameters: q is the asymmetrical Fano parameter and Γ is the width of

TABLE I. Line breadth values of the Cu $5s'\,^4D_{5/2}$ autoionization level measured at the given laser intensities. The laser wavelength was kept at 458.7 nm.

I (MW/cm^2)	Γ (cm^{-1})
1.9	17.5
0.59	11.2
0.085	8.76
0.019	6.59

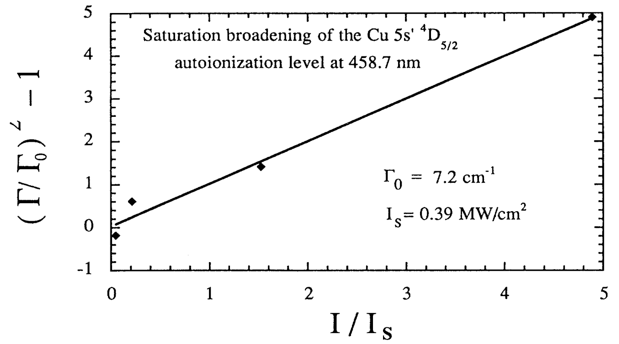

FIG. 5. Saturation broadening of the Cu $5s'\,^4D_{5/2}$ autoionization level. The solid line is the fitted saturation relation $(\Gamma/\Gamma_0)^2-1$ as a function of I/I_S, with the parameters $\Gamma_0=7.2$ cm^{-1} and $I_S=3.9\times10^5$ W/cm^2.

the autoionization state for each set of measurements. This technique is described more comprehensively in [10]. The obtained results have indicated that the linewidth parameter Γ depends on the square root of the laser intensity through the equation $\Gamma=\Gamma_0(1+I/I_S)^{1/2}$. This relation, which is described by the solid curves in Figs. 4 and 5, is known in the literature as *power broadening* or *saturation broadening* [13]. (The measured data points are presented in Table I.) It is well known that whenever saturation broadening occurs, the absorption coefficient of the optical transition κ_0 decreases according to the relation $\kappa=\kappa_0/[1+I/I_S]$, where κ is the modified absorption coefficient. It follows that the line-breadth parameter Γ depends on the laser intensity through Eq. (3):

$$\Gamma^2=\Gamma_0^2(1+I/I_S) \ . \qquad (3)$$

Based on these assumptions, the optogalvanic line profiles are broadened whenever saturation occurs. A plot of the measured linewidth as a function of the laser intensity is presented in Fig. 4. From the solid curve that is presented in Fig. 4, it is possible to evaluate the natural linewidth of the autoionization level Γ_0 and the saturation intensity I_S. In our case, the results are $\Gamma_0=7.18\pm0.59$ cm^{-1} and $I_S=0.39\pm0.09$ MW/cm^2. These two values were used to construct Fig. 5. The HCD tube filled with 5 torr of neon operated at currents below 20 mA. The pressure and Stark broadenings are less than the Doppler broadening, which is about 0.07 cm^{-1}, below the spectral resolution of our measurements.

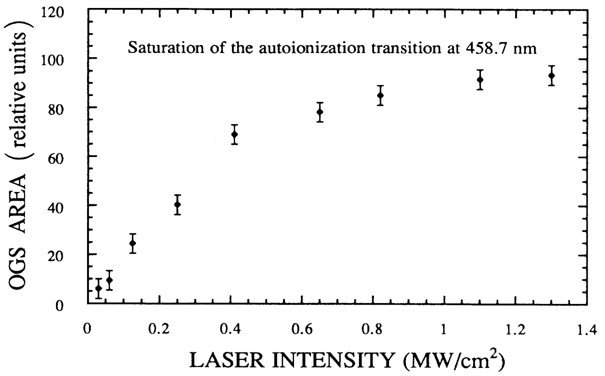

FIG. 6. Saturation curve measured at 458.7 nm laser wavelength for the Cu $5s'\,^4D_{5/2}$ autoionization level. The data points are the measured optogalvanic-signal areas at different laser intensities.

The radiative linewidth of the Cu $5s'\,^4D_{5/2}$ autoionization level was measured by Kerkhoff *et al.* [14] using an ionization cell; that value is 6.6 cm^{-1}, comparable to our measured value for the same autoionization level.

The saturation intensity was tested by another method. In this method, the dye-laser wavelength is locked at the resonance of the autoionization transition $4p'\,^4F^{\circ}_{7/2}\rightarrow5s'\,^4D_{5/2}$ (at 458.7 nm), and neutral-density filters are used to change the dye-laser intensity. The temporal integrated area of each recorded optogalvanic signal was measured and sketched as a function of the laser intensity. Here we got the familiar saturation curve. On this curve, saturation of the laser intensity can be estimated. This saturation curve is demonstrated in Fig. 6. Here the saturation intensity is about 0.4 MW/cm^2, which is in quite good agreement with the value measured in the first case.

In summary, we have directly measured the saturation broadening of the autoionization level by optogalvanic spectroscopy. Also, we have determined the Fano parameters for the Cu autoionization level, as well as power saturation for this level. In this sense, this method is applicable to spectroscopy of levels interacting with the continuum states under strong laser fields.

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