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HIGH FREQUENCY STARK EFFECT IN HELIUM I AND ITS APPLICATION IN PLASMA DIAGNOSTICS*

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We present an improved method for calculating the satellite pattern produced by the high frequency Stark effect in He I which is good for arbitrary frequency and field strength. Good agreement is obtained with experimental measurements on the 4388 and 4922 He I lines under conditions for which predictions based on previous theories fail.

The high-frequency Stark effect in He I has been proposed as a nonperturbing spectroscopic means of determining the frequency and intensity of oscillating electric fields in plasmas,[1] and has been used for this purpose by several authors.[2-9] The first calculations of the Stark patterns of He I lines due to high-frequency electric fields were made by using second-order time-dependent perturbation theory, and considered only two interacting upper levels.[1,10] For low field strengths, this pattern consists of the strong allowed line plus two "satellites" of an adjacent forbidden line, and has been shown to agree with the measured pattern for the 4388 Å He I line ($5^1D \rightarrow 2^1P$) with $E_{rms} \approx 220$ V/cm and $\nu = 31.6$ GHz.[3]

Recent experiments in a fast θ-pinch plasma[2,9] have indicated the possible disappearance of one of the two satellites for sufficiently large values of the electric field strength (estimated to be at least 7 kV/cm in the second of these two experiments), which would imply a breakdown of the perturbation calculations and the need for an improved theory for the method to be truly useful in plasma diagnostics. Two attempts have been made to improve the perturbation calculations; Kunze et al. introduced a phenomenological damping constant,[2] and also extended the perturbation calculations to the next higher order,[9] but the fundamental limitation of a perturbation-type calculation remained. We have recently applied a method due to Autler and Townes[11] to calculate the high-frequency Stark patterns for the 4922 Å line ($4^1D \rightarrow 2^1P$) and also for the 4388 Å line ($5^1D \rightarrow 2^1P$) of He I.[8] This method, a series solution of the time-dependent Schrödinger equation, also used only two upper levels, but did include Stark shifts and higher-order satellites, important effects in strong electric fields which had been neglected in previous theories. Agreement with measured patterns ($E_{rms} = 3.54$ kV/cm; $\nu = 35.2$ GHz) was good for the 4922 line, but very poor for the 4388 line. We have now extended these calculations to include all significant interacting upper levels, and find very good agreement with the measured patterns even for the 4388 line.

Following Autler and Townes,[11] we start from the time-dependent Schrödinger equation of an atom in a time-varying electric field $\underline{E}_0 \cos \omega t$,

$$i\hbar\dot{\psi} = H(t)\psi, \qquad H(t) = H_0 - \underline{\mu}\cdot\underline{E}_0 \cos \omega t \tag{1}$$

and assume that ψ is of the form

$$\psi = \sum_{\beta=1}^{N} T_\beta(t) U_\beta; \quad U_\beta \text{ satisfies the eigenvalue equation } H_0 U_\beta = (\hbar\omega_\beta) U_\beta .$$

If we further assume a solution of the form

$$T_\beta(t) = e^{-i\lambda t} \sum_{k=-\infty}^{\infty} t_{\beta k} e^{-ik\omega t},$$

Eq. (1) yields

$$(-\lambda + \omega_\beta - k\omega) t_{\beta k} = - \sum_{\alpha=1}^{N} B_{\beta\alpha} t_{\alpha k+1} - \sum_{\alpha=1}^{N} B_{\beta\alpha} t_{\alpha k-1} ;$$

$$\beta = 1, \cdots, N; \qquad k = -\infty, \cdots, -\infty. \tag{2}$$

Here $B_{\beta\alpha} = - \langle U_\beta | \underline{\mu} \cdot \underline{E}_0 | U_\alpha \rangle / 2\hbar$, and is proportional to the electric dipole moment between states β and α, λ is a constant to be determined, and N is the number of upper levels considered. With N = 2, Eq. (2) is just Eq. (7) of Autler and Townes. They derive a solution for their form of this equation in terms of an infinite continued fraction. We solve Eq. (2) for arbitrary N by first assuming $t_{\alpha k} = 0$ for $|k| > K$. Equation (2) is then a finite set of linear homogeneous equations and can be posed as an eigenvalue problem:

$$\underset{\sim}{\underline{A}}\underline{x} = \lambda \underline{x} , \tag{3}$$

where $\underline{x}$ is a column vector made up of the $t_{\alpha k}$'s. We have used a

CDC 6600 computer to solve Eq. (3) numerically for specific values of E_0 and ω.

The interpretation of λ and the $t_{\alpha k}$'s is discussed by Autler and Townes. Briefly, λ is closely related to the Stark shift of a level, and, together with the frequency ω, determines the positions of the satellites; their intensities are proportional to $(t_{\alpha k})^2$.

To obtain the "no field" wave functions $\{U_\beta\}$ we make the Coulomb approximation for He. Thus we assume the He eigenfunctions are adequately represented by one electron in the ground state and the other in a hydrogenic excited state described by quantum numbers n, ℓ, m. For the energy of a given n, ℓ state we have used values given by Martin.[12] We have studied transitions in the vicinity of two allowed lines, which we will call

Case I: 4922 Å (4^1D, etc. $\rightarrow 2^1P^o$)

and Case II: 4388 Å (5^1D, etc. $\rightarrow 2^1P^o$).

In the presence of an electric field the upper and lower states are modified due to coupling to other eigenstates by the electric dipole matrix element in Eq. (2). The coupling is strongest to nearby states, and becomes insignificant for energy levels separated by more than about 100 $\hbar\omega$. The electric dipole moment operator does not couple singlet and triplet states, hence, in considering the above singlet transitions we have ignored all triplet states. For Case I we limit the $\{U_\beta\}$ to the set of hydrogenic wave functions with $n = 4$, $\ell = 0$ to 3, and $|m| \leq \ell$. For Case II we limite the $\{U_\beta\}$ to the set $n = 5$, $\ell = 0$ to 4, and $|m| \leq \ell$. In all cases that we consider, the effect of the electric field on the $2P^o$ state is negligible.

In our experiment the electric field is linearly polarized and can be assumed without loss of generality to lie in the z direction. With this choice the electric dipole moment operator does not couple eigenstates with different m values, and the matrix A reduces to an especially simple form to diagonalize. We have set $K = 10$; this insures adequate numerical accuracy for these two cases.

We generate a high-frequency electric field in a cylindrical microwave cavity and apply it to a He plasma produced by a dc discharge in a 1/3-mm-i.d. quartz capillary which threads the cavity. The field frequency is much higher than either the plasma frequency or the electron collision frequency, so that the microwave field has no noticeable effect on the plasma. Further details of the experimental arrangement are given in Ref. 3. Two changes should be noted, however. The cavity is now excited at 35.2 GHz by a 10-watt CW klystron. With the higher power and higher electric field the satellites are no longer buried in the wings of the allowed line; this permits the experiment to be run CW and obviates the need for phase-sensitive detection. The light intensity at a given wavelength is now measured by use of standard photon-counting techniques.

For a direct comparison of the theoretical calculations with the measured line profiles, we have folded the theoretical results, which consist of a discrete line spectrum, with a realistic "instrument function" obtained from measurements taken on the same apparatus but with the microwave power turned off. Most of the observed broadening was instrumental. Figures 1 and 2 show comparisons of experimental results with various theories, all calculated for observation at right angles to the direction of the field and for a peak field strength of 5.0 kV/cm (3.54 kV/cm rms). In all cases $\Delta\lambda = 0$ is the position of the allowed line in the absence of the perturbing electric field. All "bumps" on the theoretical profiles are produced by one or more satellites and not by irregularities in the instrument function. All satellites stronger than 10^{-5} of the total intensity of the pattern were retained in the calculations.

Figure 1 shows a comparison between experimental and theoretical results for Case I. The multilevel theory outlined above, the Autler-Townes theory, and the perturbation theory of Baranger and Mozer all give nearly the same results for the predicted spectrum; the major discrepancy between them comes from the neglect of the Stark shift in the perturbation calculation. The slight difference between the Autler-Townes and the multilevel theories is due to the retention of the 4P level in the latter. In both these cases we have retained 18 satellites. Agreement of the multilevel theory with experiment is excellent, and even the other two theories agree quite well with experiment for this field strength. The close agreement of the perturbation calculations and the multilevel calculations, which we expect to be much more accurate, indicate that the perturbation calculations can still be trusted for this line at this frequency and field strength, and we may in fact use them to determine the field strength. The value so obtained was 5 kV/cm peak field with an estimated error of less than 500 V/cm.

Case II, shown in Fig. 2, is a much more severe test of the various theories because

(a) the Stark shift increases with n, hence the effect of a given E field is greater on the 4388 line,

(b) the energy levels of n = 5 are closer together, so that more satellites (i.e., higher-order transitions) become important, and

(c) for n = 5 there is a G level very near the F level, and the two interact strongly.

In Fig. 2 we compare the measured line profile for the 4388 line with theoretical ones calculated from our multilevel theory and from the Autler-Townes theory, using the field strength derived from the measurements on the 4922 line. Agreement between the multilevel calculations and the measured data is very good, whereas experiment and the Autler-Townes calculations sharply disagree, not only in satellite positions and intensities but also in the Stark shift of the allowed line. This disagreement graphically illustrates the need to include additional upper levels, as this is the only significant difference in the two theories. Perturbation calculations, not shown, disagree even more strongly with the measurements. In the Autler-Townes calculations we include 42 satellites; 58 were used in the multilevel calculations. Remaining discrepancies between the measured data and the multilevel calculations may be due to errors in the energy levels used in the calculations.

In using the high-frequency Stark effect to measure oscillating electric fields in a plasma, it is very desirable to be able to use the simple perturbation calculations of Baranger and Mozer, if possible, rather than the much more accurate but laborious machine calculations as we have done. In many practical cases the Baranger-Mozer theory is sufficiently accurate. We have calculated a limited number of high-frequency Stark patterns for the 4388 and the 4922 lines of He I for various values of E and ν in order to try to estimate the range of applicability of the Baranger-Mozer theory for these two lines.

For weak electric fields, a "normal" high-frequency Stark pattern, consisting of the allowed line ($n^1D \rightarrow 2^1P$) and two satellites of the forbidden line ($n^1F \rightarrow 2^1P$), is observed; the satellite intensities are accurately predicted by perturbation calculations. These two satellites are usually identified as "far" or "near," according to their proximity to the allowed line. If we increase the electric field strength, keeping the frequency constant, a number of changes not predicted by second-order perturbation theory occurs. The intensities of both satellites grow, but at different rates, other satellites appear (higher-order transitions from the F and from other angular momentum states to the 2P state), and increasingly important Stark shifts cause the different groups of satellites to shift relative to each other and to the allowed line. The positions and intensities of the satellites change very rapidly near a resonance (a condition in which the separation of two upper levels, including the Stark shifts, is an integral multiple of the field frequency), but this effect probably will not normally be observable in a plasma experiment because of inadequate resolution. In the limit of very high field strengths ($E_{rms} \gg 20$ kV/cm for the 4922 line), Stark shifts exceed the separation of the unperturbed energy levels, and one can use the calculations by Blochinzew[13] for hydrogen-like atoms.

Provided the upper levels are populated according to their statistical weights, the second-order perturbation theory seems capable of calculating the ratio of the intensity of the far satellite to that of the allowed line with an error of less than 20% as long as the intensity of the far satellite does not exceed that of the near satellite, a convenient measure of the breakdown of the perturbation calculations. Under these conditions, other satellites of comparable intensity are typically also present in the pattern. For the 4922 line with $\nu = 35.2$ GHz, this limit is reached at $E_{rms} \approx 10$ kV/cm. The intensity of the near satellite is less reliable as an indicator of electric field strength, as was pointed out in Ref. 9.

For field strengths exceeding a few kV/cm, the Stark shifts of the various levels must be taken into account in calculating the positions of the satellites. As an example of the importance of this effect, we cite a recent publication in which the frequency of an electric field presumed present in the plasma was calculated from the observed position of a satellite near the 4922 line (assumed to be the far satellite of the forbidden line) relative to the position of the allowed line.[9] Stark shifts of the two lines, which tend to separate them, were ignored, although for the conditions quoted ($E_{rms} \geq 7$ kV/cm) the net Stark shift would produce an apparent displacement of the satellite at least double that which was observed and equated to the field frequency.

The simplest and safest way to apply the high-frequency Stark effect to measure strong electric fields in plasmas, if one does not wish to carry out the numerical calculations outlined above, is to use the observed separation of satellites originating from a particular angular momentum state to determine the frequency of the electric field (this separation is always twice the field frequency, independent of the strength of the field), and to use the observed Stark shifts to determine the strength of the electric field. No knowledge is then required of the distribution of excited states. Numerous computer calculations for the 4922 line indicate that the Stark shifts are essentially independent of frequency unless one is near a low-order resonance. Since the 4922 line is a particularly useful line for plasma diagnostics, we describe below a simple method for using it to determine the electric field strength.

A convenient quantity to measure is the separation of the (shifted) allowed line from the average of the positions of the far and near satellites. The net Stark shift Δ we define to be this separation minus 1.337 Å, which is the separation in the absence of an electric field. Approximate expressions for Δ for the 4922 line, in Å, as a function of the rms electric field strength in kV/cm, are

$$\Delta \approx 0.0095\, E_{rms}^{2}\ \text{Å}, \qquad E_{rms} < 3\ \text{kV/cm, and}$$

$$\Delta \approx 0.0133\, E_{rms}^{1.71}\ \text{Å}, \qquad 3 < E_{rms} < 10\ \text{kV/cm.}$$

FOOTNOTES AND REFERENCES

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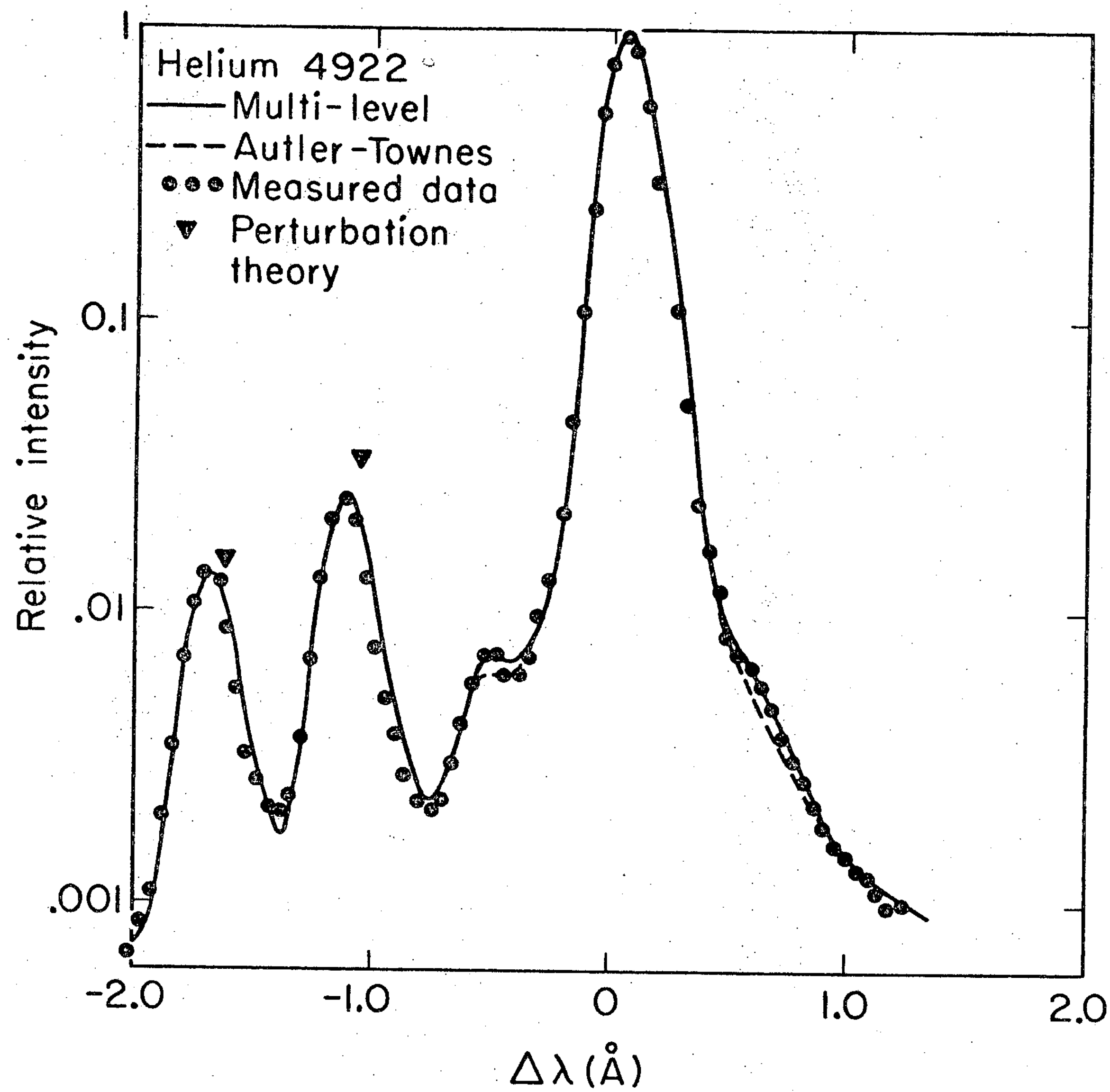
FIGURE LEGENDS

Fig. 1. Case I, (4^1D, etc. → 2^1P^o) 4922 Å He I.

Fig. 2. Case II, (5^1D, etc. → 2^1P^o) 4388 Å He I.



XBL703-2461

Fig. 1

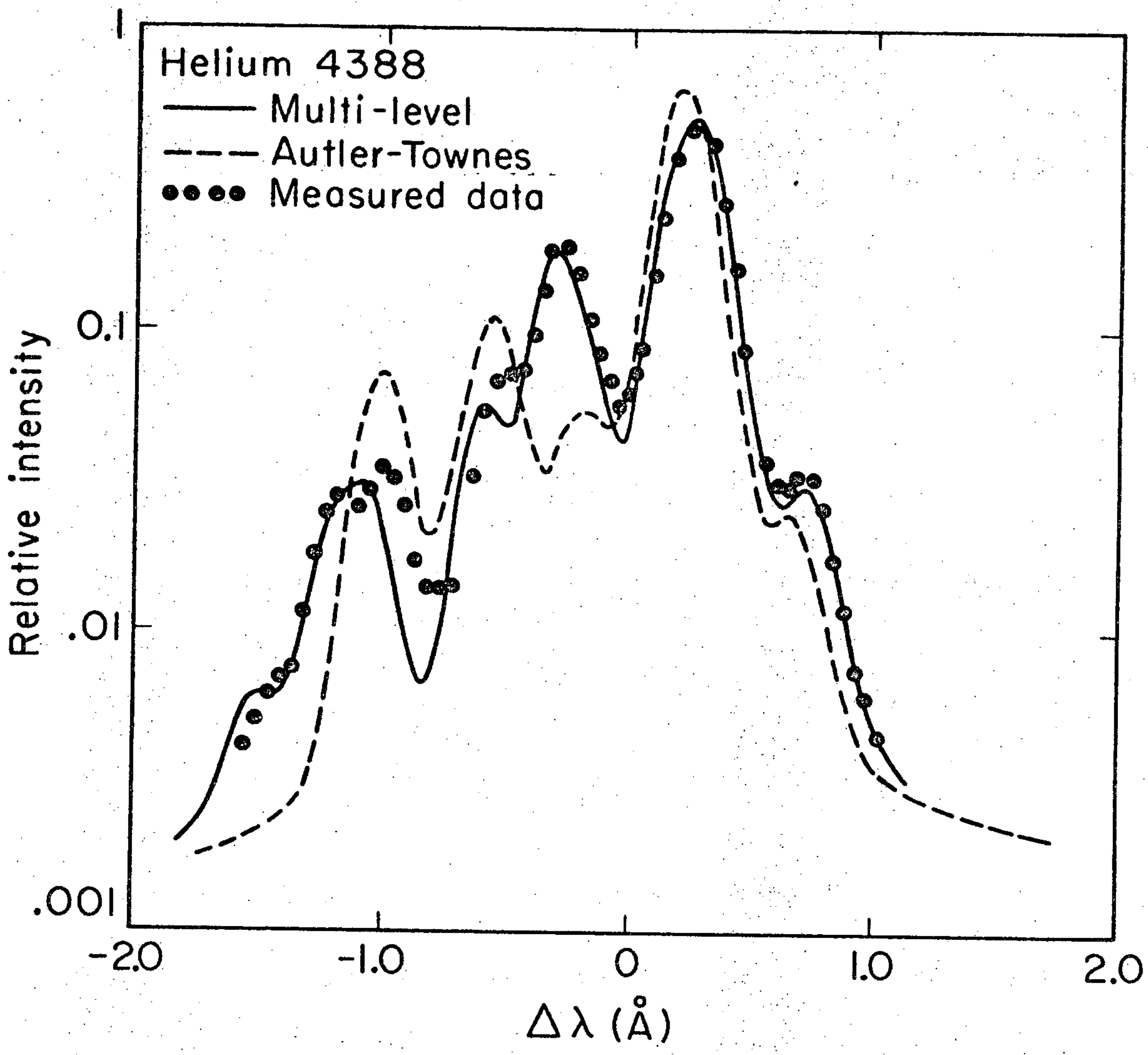

Helium 4388
Multi-level
Autler-Townes
Measured data
Relative intensity
1
0.1
.01
.001
-2.0
-1.0
0
1.0
2.0
Δλ (Å)
XBL703-2460

Fig. 2

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