# Light as a Plasma Probe*†

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(Received February 27, 1961)

The effect of longitudinal plasma oscillations on atomic spectra is examined. It is shown that they should give rise to satellite lines, disposed symmetrically in pairs about a forbidden line and separated from it by $\Omega_p$, the plasma frequency. Discussion is given of the circumstances under which these satellites should be strong enough to be observed. Their observation would amount to a measurement of the frequency and intensity of plasma oscillations.

SPECTROSCOPIC observations of the light emitted by ionized gases can be used to determine various properties of plasmas, such as the ion density[1] and the temperature.[2] Should there also be observable spectroscopic effects of longitudinal plasma oscillations?[3] In the present paper, this question will be answered by the affirmative. It will be shown that, when plasma oscillations occur in a cool plasma, i.e., one containing atoms or ions emitting spectral lines, they should give rise to satellite lines in the spectrum. The authors are unaware of any experiment specifically designed to observe this effect, although its existence might even be revealed by data already obtained. It is hoped that this paper will provide stimulation for someone to attempt the observation. The effect constitutes essentially a new plasma probe which should permit, under certain conditions, a measurement of the frequency and the intensity of the oscillations.

## THEORY

The simplest way to visualize the production of the satellites is as the result of a second-order transition involving the plasma field and the emission of light. Figure 1 shows three energy levels of an atom involved in such a transition. Hydrogenlike atoms are not suitable, because of their large Stark effect, so one could consider helium, for example. In Fig. 1, $A$ denotes an allowed transition and $F$ a forbidden transition for electric dipole radiation. The atom is assumed to be initially in state $l'$.[4] Under the influence of the plasma field, it undergoes transition 1 to state $l$. This is followed by a radiative transition 2 to the final state $l_0$. The intermediate state $l$ is virtual, i.e., energy is not conserved and the atom remains there for a short time only. But energy must be conserved in the over-all process, and since the plasma oscillation transfers the energy $\pm\hbar\Omega$ to the atom, where $\Omega$ is the frequency of the oscillation, the frequency of the emitted light is $F\pm\Omega$, where $F$ is the frequency of the forbidden line. Thus there should appear in the spectrum two lines separated from the forbidden line by frequency $\Omega$, as shown in Fig. 2.

The strength of the satellites can be easily calculated by second-order time-dependent perturbation theory.[5] The interaction giving rise to transition 1 is $-\mathbf{d}\cdot\mathbf{E}$, $\mathbf{E}$

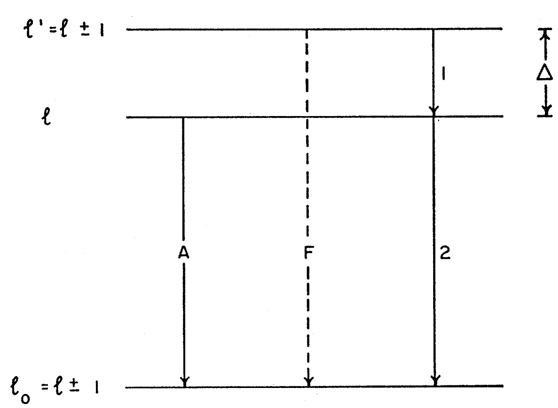


FIG. 1. Three energy levels of a non-hydrogenlike system. The two upper states have the same principal quantum number and are very close. The transition frequencies to the lower state are in the optical region.

* Support was received from the Office of Naval Research, the Atomic Energy Commission, and the joint General Atomic—Texas Atomic Energy Research Foundation program on controlled thermonuclear reactions.

† This work was reported at the annual meeting of the American Physical Society, Division of Plasma Physics, in Gatlinburg, Tennessee, November, 1960.

[1] H. R. Griem, A. C. Kolb, and K. Y. Shen, Phys. Rev. **116**, 4 (1959); B. Mozer, Ph.D. thesis, Carnegie Institute of Technology, 1960 (unpublished); H. R. Griem, M. Baranger, A. C. Kolb, and G. Oertel (to be published); and further work in preparation.

[2] W. Wiese, H. F. Berg, H. R. Griem, Phys. Rev. **120**, 1079 (1960).

[3] Since the original suggestion of L. Tonks and I. Langmuir [Phys. Rev. **33**, 195 (1929)], oscillations that seem to behave like electrostatic plasma oscillations have been observed many times. Among the recent work, see for instance D. H. Looney and S. C. Brown, Phys. Rev. **93**, 965 (1954); G. D. Boyd, L. M. Field, and R. W. Gould, Phys. Rev. **109**, 1393 (1958); D. W. Mahaffey, J. Electron Contr. **6**, 193 (1959); I. F. Kharchenko, Ya. B. Fainberg, R. M. Nikolaev, E. A. Kornilov, and N. S. Pedenko, Zhur. Eksptl. i Teoret. Fiz. **38**, 685 (1960) [translation: Soviet Phys.—JETP **11**, 493 (1960)].

[4] For simplicity, we designate states by their orbital angular momentum quantum numbers.

[5] L. I. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1949), 1st ed., Chap. 8.

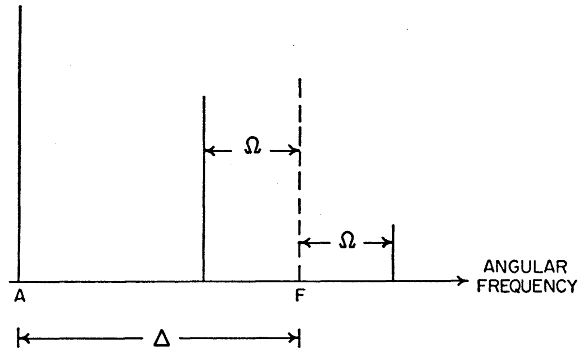


FIG. 2. Spectrum of radiation emitted by an atom with the energy levels pictured in Fig. 1 and perturbed by a plasma field oscillating with frequency Ω.

being the oscillating plasma field and **d** the dipole moment of the atom, and one uses the usual[6] electromagnetic interaction for the second part of the transition. The strength (i.e., the integrated intensity) of the satellites in units of the strength of the allowed line is found to be

$$S=\hbar^2\langle E_p{}^2\rangle \mathfrak{R}_{ll'}/6m^2e^2(\Delta\pm\Omega)^2, \tag{1}$$

where $m$ and $e$ are the mass and charge of the electron, $\Delta$ is the splitting (in angular frequency units) between the allowed and the forbidden line, $\langle E_p{}^2\rangle$ is the time-average of the plasma field squared, and $\mathfrak{R}_{ll'}$ is a dimensionless radial integral,[7]

$$\mathfrak{R}_{ll'}=\frac{\max(l,l')}{2l+1}\left[\frac{\int_0^\infty R_l(r)R_{l'}(r)r^3dr}{a_0}\right]^2, \tag{2}$$

with $a_0=\hbar^2/me^2$. The stronger satellite is the one closer to the allowed line. The appearance of the resonance denominator $\Delta\pm\Omega$ is expected since, for $\Delta=\pm\Omega$, the plasma field would have just the right frequency to induce transition 1 of Fig. 1 as a real transition. However, Eq. (1) is not valid right at resonance, since perturbation theory has been used; it is restricted to cases where $S$ turns out to be small compared to unity.

Of course, the effect just discussed is none other than time-dependent Stark effect. If the plasma field were time-independent, it would produce a forbidden line through static Stark effect in the well-known manner. The excitation of the forbidden line may also be viewed as the result of a second-order transition: The static field causes transition 1, then the electromagnetic interaction causes transition 2. The only difference is that, in the present case, the field is not static so that energy $\pm\hbar\Omega$ is transferred to the radiation, causing a displacement of the forbidden line by the same amount; the resonance denominator is correspondingly changed also. But, for a static field, Eq. (1) is actually identical to the result of ordinary Stark effect theory. As the frequency of the field is increased, the forbidden line splits into these two components that move away from each other,[8] and their intensity changes because of the modified resonance denominator.

The perturbation treatment says nothing about the shape of the satellites. Obviously, they will always be wider than the allowed line. All effects that broaden the allowed line, Doppler broadening, and pressure broadening, will also broaden the satellites. But, in addition, the satellites have some broadening causes of their own: damping of plasma oscillations and spatial inhomogeneities which result in a spread in the frequency Ω. To make a complete theory of the line shape, and also to treat the near resonant case when Eq. (1) does not give a small result, it is necessary to call upon the general line-shape formula, according to which the Fourier transform $\Phi(s)$ of the spectrum $F(\omega)$ is given by[9]

$$\Phi(s)=\int_{-\infty}^{\infty}\exp(-i\omega s)F(\omega)d\omega$$
$$=[\langle lm|U(s)|lm\rangle]_{\text{av}}. \tag{3}$$

Here $|lm\rangle$ is one of the substates of $l$ and $U(s)$ is the interaction representation evolution operator which obeys

$$i\hbar dU/ds=\exp(iH_0s/\hbar)V(s)\exp(-iH_0s/\hbar)U(s), \tag{4}$$

where $H_0$ is the unperturbed Hamiltonian and $V(s)=-\mathbf{d}\cdot\mathbf{E}(s)$ the perturbation. The average is over all possible electric fields $\mathbf{E}(s)$ and also over the magnetic quantum number $m$. The normalization has been chosen such that $\Phi(s)$ is unity, and therefore $F(\omega)$ is a $\delta$ function, when there is no perturbation. The frequency of the unperturbed allowed line is taken as origin. With Eq. (3), one can compute the whole spectrum in the vicinity of $A$ and $F$, one can show that there are some additional weaker satellites besides the two we have mentioned, one can compute the reduction in intensity of the allowed component which is necessary to compensate for the appearance of satellites, and one can treat the case of resonance. It seems premature to go into these questions at this time, hence it will only be shown here that Eq. (3) yields back Eq. (1) when the plasma field can be treated as small.

To do this, we assume that the electric field is of the

[6] W. Heitler, *The Quantum Theory of Radiation* (Oxford University Press, New York, 1944), 2nd ed.

[7] This is the integral that comes in the calculation of the oscillator strength. For an alternative expression, see Eq. (9). It will usually be sufficient to use the hydrogenic approximation given by H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-Electron Atoms* (Academic Press, Inc., New York, 1957), Eq. (63.5).

[8] Actually second-order effects split the forbidden line even when the field is static. Similar perturbations of the spectrum calculated here will occur for time-dependent fields, but they will usually be small and completely masked by pressure broadening.

[9] P. W. Anderson, Phys. Rev. **76**, 647 (1949). The notation is that of M. Baranger, Phys. Rev. **111**, 494 (1958). Various simplifying assumptions have been made to obtain Eq. (3) of this paper from Eq. (13) of the latter reference, namely: neglect of the interaction in the final state (one-state case); there are no two initial states with the same angular momentum and parity quantum numbers (no overlapping lines); all initial states are equally likely ($kT\gg\hbar\Delta$).

from

$$\mathbf{E}(s)=\mathbf{E}_p\cos(\Omega s+\varphi). \quad (5)$$

Besides this plasma field, which is generated by the long-range correlations between the electrons, there are also random fields due to the electrons and ions in the immediate vicinity of the light-emitting atom. These latter fields give rise to pressure broadening[10]; they are neglected in the present simplified treatment. One expands $U(s)$ to second order. The zeroth order term gives the unperturbed allowed line. The first-order term averages to zero. The second-order term in $\Phi(s)$ is

$$\Phi^{(2)}(s)=-\hbar^{-2}\sum_i\int_0^s dt\int_0^t dt'\exp[i(\omega_l-\omega_i)(t-t')] \times[\langle lm|V(t)|i\rangle\langle i|V(t')|lm\rangle]_{\text{av}}. \quad (6)$$

Here the intermediate states $i$ consist of the magnetic substates of level $l'$, hence one obtains (using Greek subscripts for Cartesian components)

$$\Phi^{(2)}(s)=-\hbar^{-2}\sum_{m'\alpha\beta}\int_0^s dt\int_0^t dt'\exp[i(\omega_l-\omega_{l'})(t-t')] \times\langle lm|d_\alpha|l'm'\rangle\langle l'm'|d_\beta|lm\rangle \times[E_{0\alpha}E_{0\beta}\cos(\Omega t+\varphi)\cos(\Omega t'+\varphi)]_{\text{av}}. \quad (7)$$

The average over the phase, the direction,[11] and the amplitude of the field is

$$\tfrac{1}{6}\delta_{\alpha\beta}\langle E_p^2\rangle\{\exp[i\Omega(t-t')]+\exp[-i\Omega(t-t')]\}. \quad (8)$$

The sum over $\alpha$ and $m'$ of the dipole matrix elements is essentially the quantity $\mathcal{R}_{ll'}$ defined by Eq. (2),

$$\textstyle\sum_{\alpha m'}|\langle lm|d_\alpha|l'm'\rangle|^2=e^2a_0^2\mathcal{R}_{ll'}. \quad (9)$$

The time integrals are elementary and one finds

$$F^{(2)}(\omega)=\frac{e^2a_0^2\mathcal{R}_{ll'}\langle E_p^2\rangle}{6\hbar^2\omega^2}[\delta(\omega+\omega_l-\omega_{l'}+\Omega) +\delta(\omega+\omega_l-\omega_{l'}-\Omega)], \quad (10)$$

i.e., the same spectrum as in Eq. (1) and Fig. 2.

## DISCUSSION

One might think of using the effect to try to detect thermal plasma oscillations, i.e., the situation where each possible mode of vibration has average energy $kT$. Since the vibration frequency depends on wave number,[12] this would give rise not to fairly sharp satellites, but to two bands extending out from the Langmuir frequency

$$\Omega_p=(4\pi ne^2/m)^{\frac{1}{2}}. \quad (11)$$

The shape of these bands can be computed from Eq. (10) and the knowledge of the dispersion law[12] for plasma oscillations. Their intensity must then be compared with that of the pressure-broadened tail of the allowed line,[10] which acts as background. In typical cases, one finds that the plasma band would be only 10 or 20% of the background. Since various broadening effects prevent the onset of the band from being sharp, it would seem that, with present techniques, such bands would be very hard to detect.

Hence the best chance of observing the effect under discussion here lies in nonequilibrium situations such as those realized in the experiments[13] mentioned in reference 3. One needs to excite a strong level of plasma oscillations in a small range of frequencies,[14] which would presumably be close to the Langmuir frequency (11). The narrower the range of frequencies, the sharper the satellites will be, and consequently the easier their detection. It will be seen in the following that, if their width can be made of the same order of magnitude as that of the allowed line, observation of the satellites is definitely in the realm of possibility.

The integrated intensity in each satellite is given by Eq. (1). To illustrate its use, the values of $\Delta$ and $\mathcal{R}_{ll'}$ for a few helium lines are listed in Table I. To conform with spectroscopic practice, $\Delta$ is given in wave number units,[15] $\text{cm}^{-1}$. In the same units, the plasma frequency $\Omega_p$ equals approximately 1 $\text{cm}^{-1}$ for an electron density $n$ of $10^{13}$ $\text{cm}^{-3}$ and varies proportionally to $n^{\frac{1}{2}}$. Hence it would seem that, to make possible a spectroscopic measurement of $\Omega_p$, $n$ would have to be at least $10^{13}$ $\text{cm}^{-3}$. At lower densities, the width of the lines would interfere with the accuracy of the measurement. However, at these low densities, it might still be possible to detect the existence of plasma oscillations, though not their frequency, through their excitation of the for-

TABLE I. The splitting $\Delta$ between allowed and forbidden line,[a] in $\text{cm}^{-1}$, and the dimensionless quantity $\mathcal{R}_{ll'}$ of Eqs. (2) and (9),[7] for various lines of helium. Other notations as in text.

| Wavelength (in angstroms) | $l$ | $l'$ | $\Delta$ | $\mathcal{R}_{ll'}$ |
|---|---|---|---|---|
| 5016 | $3\,^1P$ | $3\,^1D$ | 104 | 67 |
| 6678 | $3\,^1D$ | $3\,^1P$ | 104 | 40 |
| 3188 | $4\,^3P$ | $4\,^3D$ | 227 | 288 |
| 4472 | $4\,^3D$ | $4\,^3F$ | 8 | 151 |
| 3965 | $4\,^1P$ | $4\,^1D$ | 46 | 288 |
| 4922 | $4\,^1D$ | $4\,^1F$ | 6.5 | 151 |
| 2945 | $5\,^3P$ | $5\,^3D$ | 116 | 787 |
| 4026 | $5\,^3D$ | $5\,^3F$ | 4.5 | 540 |
| 3614 | $5\,^1P$ | $5\,^1D$ | 24 | 787 |
| 4388 | $5\,^1D$ | $5\,^1F$ | 1.8 | 540 |
| 2829 | $6\,^3P$ | $6\,^3D$ | 67 | 1728 |
| 3448 | $6\,^1P$ | $6\,^1D$ | 14 | 1728 |

[a] Landolt-Börnstein, *Zahlenwerte und Funktionen* (Springer-Verlag, Berlin, 1950), Vol. 1, Part 1.

[10] H. R. Griem, M. Baranger, A. C. Kolb, and G. Oertel (to be published).

[11] It is assumed here that the direction of the plasma field is random. This may not always be the case. If the plasma field oscillates predominantly in one direction, the satellites will be emitted nonisotropically and will be polarized, but expression (1) or (10) will still be valid for the integrated intensity. This is because one can replace the average over directions of $\mathbf{E}_p$ by an average over $m$ without changing the result. If the direction is random, the average over $m$ is not necessary.

[12] A. Vlasov, Zhur. Eksptl. i Teoret. Fiz. 8, 291 (1938); D. Bohm and E. P. Gross, Phys. Rev. 75, 1851 (1949).

[13] However, as will be seen shortly, the electron density $n$ would have to be appreciably higher than in the experiments of reference 3.

[14] This point was first stressed by S. Cunningham in a private conversation for which the authors are much indebted.

[15] Multiply by $2\pi c$ to convert to angular frequency.

bidden line.[16] That is, one is now talking about ordinary, time-independent Stark effect. Since the random fields of the plasma[17] already excite the forbidden line, it would be necessary that the plasma field be larger than these random fields.

It is seen from Table I that, by choosing an appropriate line, one can always make $\Omega_p$ and $\Delta$ of the same order of magnitude. It might be thought desirable to have $\Omega_p$ and $\Delta$ as nearly equal as possible in order to achieve resonance. But this is not the case, because the satellite must be observed on the background of the tail of the allowed line, and this tail decreases like the inverse square frequency or faster, so that the ratio of satellite to background stays the same or improves as one goes off resonance. In fact, if the spectrum is very clean, one might want to look for the weaker satellite on the other side of the forbidden line.

In order to estimate the relative spectral intensities of satellite and background, one can assume that the allowed line has a Lorentz shape

$$I_A=\pi^{-1}w_A(\omega^2+w_A^2)^{-1}, \tag{12}$$

where $w_A$ is the half-width (half the width at half-maximum). At the position of the satellite, $\omega=\Delta\pm\Omega$ so that, if one is far from resonance, one has

$$I_A\approx\pi^{-1}w_A(\Delta\pm\Omega)^{-2}. \tag{13}$$

If the half-width of the satellite is called $w_s$, Eq. (1) gives for the satellite intensity at its center

$$I_s=\frac{1}{\pi w_s}\frac{\hbar^2\langle E_p^2\rangle\mathcal{R}_{ll'}}{6m^2e^2(\Delta\pm\Omega)^2}, \tag{14}$$

and the ratio of satellite to background is

$$\frac{I_s}{I_A}=\frac{\hbar^2\langle E_p^2\rangle\mathcal{R}_{ll'}}{6m^2e^2w_Aw_s}. \tag{15}$$

If $\Omega$ is small compared to $\Delta$, it may be more relevant to compare the strength of the satellite $S_s$ with that of the forbidden line, $S_F$. This is easy because Eq. (1) is also applicable to the forbidden line, provided it be multiplied by a factor 2 since, for $\Omega=0$, the two satellites coalesce into a single forbidden line. Then the field should be the random electric field $E_R$, which is of order[17] $2.6en^{\frac{2}{3}}$. Hence the strength ratio between a satellite and the forbidden line is simply

$$\frac{S_s}{S_F}=\frac{1}{2}\frac{\langle E_p^2\rangle}{E_R^2}\frac{\Delta^2}{(\Delta\pm\Omega)^2}. \tag{16}$$

One possible cause of width is pressure broadening. This can now be reliably computed[10] and gives rise to half-widths of the order of 1 to 10 cm$^{-1}$ for $n=10^{16}$ cm$^{-3}$ and varying roughly in proportion with $n$. Another source of width, especially for light atoms and low densities, is Doppler effect. This gives rise to very steep wings, hence $w_A$ in Eq. (15) should consist only of the pressure width. On the other hand $w_s$ should be the complete half-width expected for the satellite, including that coming from the possible spread in the frequency $\Omega$. It might be thought that one could improve the ratio (15) by picking a line with large $\mathcal{R}_{ll'}$, but this is futile because $w_A$ is itself proportional to $\mathcal{R}_{ll'}$. Any line will do provided it be strong, in a clean region of the spectrum, and have associated with it a clear forbidden line. It is obviously desirable to try to decrease the ratio of the width of the lines to the plasma frequency. As long as pressure broadening dominates, this can be achieved by lowering the density, since $w\sim n$, but $\Omega_p\sim n^{\frac{1}{2}}$. When Doppler broadening takes over, the situation is reversed. So, it would seem that the most propitious densities for observing the effect would be those where the transition from Doppler to pressure broadening occurs, which is usually around $10^{15}$ cm$^{-3}$.

For a numerical example, one can consider the line 3965 A of helium with $n=10^{15}$ cm$^{-3}$, hence $\Omega_p=10$ cm$^{-1}$. For a temperature of 2 v, the pressure and Doppler widths are approximately equal to each other and to 0.5 cm$^{-1}$. Let it be assumed that one has succeeded in putting one thousandth of the total thermal energy of the gas in plasma oscillations, which corresponds to an effective plasma field $\langle E_p^2\rangle^{\frac{1}{2}}$ of 3300 v/cm, not an unreasonable value and one of the same order of magnitude as $E_R$. Taking 0.5 cm$^{-1}$ for $w_A$ and 2 cm$^{-1}$ for $w_s$, one finds the ratio $I_s/I_A$ of Eq. (15) equal to unity. Hence, the satellites appear to be strong enough to be observed. If the absolute intensity of the satellite is a problem (it would be very weak in the above example), one can always try to match $\Delta$ and $\Omega_p$ more precisely, thus raising the satellite and the background simultaneously.

## ACKNOWLEDGMENTS

The authors are happy to thank S. Cunningham, W. E. Drummond, and A. V. Phelps for enlightening discussions. Part of this work was performed while the first-named author was a summer consultant with the John Jay Hopkins Laboratory of General Atomic. He is most grateful to the staff of the Laboratory, in particular Dr. E. C. Creutz and Dr. D. W. Kerst, for their hospitality and stimulation.

[16] This remark is due to W. E. Drummond (private communication).

[17] J. Holtsmark, Ann. Physik **58**, 577 (1919); B. Mozer and M. Baranger, Phys. Rev. **118**, 626 (1960).