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Two-Photon Absorption and Stimulated Raman Scattering on Excited Helium Atoms in a Plasma*

C. F. Burrell and H.-J. Kunze

Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742

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By focusing simultaneously microwave radiation and the beam from a tunable dye laser into a helium plasma, we observe two-photon absorption and stimulated Raman scattering on excited helium atoms. Both effects can be used to measure high-frequency electric fields in plasmas, and we discuss the advantages of this method as compared with the standard technique where such fields are investigated in emission spectra.

High-frequency electric fields can stimulate optical transitions between quantum states of atoms and ions involving more than one quantum. The new spectra are characterized by satellite lines around allowed and forbidden spectral lines. In 1961 Baranger and Mozer[1] pointed out the relevance of this phenomenon to plasma physics and proposed these effects as a diagnostic tool to study oscillating electric fields in plasmas. The first experiments were done by Kunze and Griem[2] observing plasma electric fields and by Cooper and Ringler,[3] who in a model-type experiment generated the high-frequency electric field in a microwave cavity and applied it to a separately generated plasma. In the meantime, this very powerful diagnostic technique has been studied further not only theoretically,[4,5] but also in many experiments (for a brief review, see Bekefi[6]).

Like all spectroscopic measurements, this diagnostic technique has the inherent disadvantage that light is collected along the line of sight through the entire length of the plasma, i.e., only correspondingly averaged quantities are obtained directly. It suggests itself, therefore, to apply light-scattering techniques to this problem, which offer the great advantage of local measurements. Tunable dye lasers make this now possible, and the feasibility of such techniques has been demonstrated only recently by the authors[7] who observed the resonance scattering on excited helium atoms. The new technique is complementary to the technique of collective laser scattering[8] from a plasma because it measures the spectrum of local electric field fluctuations, whereas the latter gives the spectrum of the local electron density fluctuations. Because the upper levels of both the allowed and satellite transitions are very strongly collisionally coupled, the intensity of satellite lines are enhanced by the same factor as the intensity of the allowed line when the allowed transition is optically pumped. As will be discussed later in greater detail, further advantages are obtained by using pump light at wavelengths corresponding to the satellites: The resulting processes correspond to a two- or more-quanta absorption process and to stimulated Raman scattering. The calculation for both processes has been done by Reinheimer[9] using second-order time-dependent perturbation theory of the high-frequency Stark effect; the probabilities for these processes can be obtained, of course, also from the inverse emission processes.[1,4]

In a model-type experiment we apply laser radiation and an intense microwave pulse simultaneously to a low-temperature helium plasma. This means we investigate here the coupling of high-frequency optical photons with lower-frequency microwave photons by bound atomic electrons (in a practical case, the coupling would be with the low-frequency plasmon field). Multiquantum absorption processes in molecules and atoms have been observed in the rf and microwave region for a number of years, and recently also the infrared-microwave two-photon absorption in NH_3.[10]

Figure 1 illustrates the two processes observed. The atom is initially in the level i. The transition $i \to j$ is allowed; the transition $i \to f$ is forbidden—it corresponds to $\Delta L = 0$ or ± 2. Keeping the microwave frequency ω_M fixed, the laser frequency ω_L is now tuned (a) until $\hbar\omega_L + \hbar\omega_M$ corresponds exactly to the energy difference between the levels i and f resulting in the two-photon absorption process, or (b) until $\hbar\omega_L - \hbar\omega_M$ corresponds to this energy difference, resulting in the absorption of the optical photon and the stimulated emission of a microwave photon. Both processes increase the population of the level f and can be observed, thus, as enhancement of the intensity of all lines originating at this level. In addition, this enhancement can also be seen on lines originating at close-by levels to which the collisional transfer rate of the excitation is faster than the radiative decay rate of the level f, thus, for example, on the allowed transition $j \to i$. Since allowed $(j \to i)$ and forbidden transitions $(f \to i)$ are very close in wavelength, in practical cases it is, of course, advantageous for experimental reasons to use as the monitor line for the absorption one which is in a different wavelength region. This minimizes difficulties caused by stray light from the original laser beam.

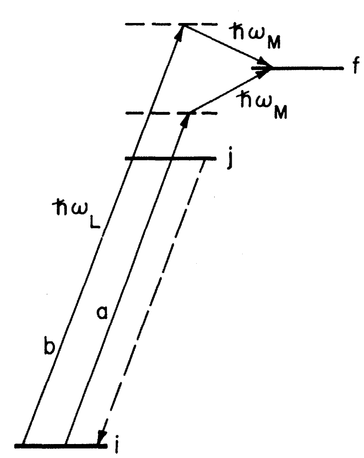

FIG. 1. Energy-level diagram illustrating the two-quantum processes observed.

In the present experiment the magnetic field is negligible, so we neglect modifications of the spectrum due to the Zeeman effect.[4] From an experimental point of view it is convenient not to measure the absorption of the satellites absolutely, but rather compare this absorption with the absorption of the nearby allowed transition $i \to j$. The ratio $S_\pm$ of the absorption coefficient at the satellite wavelength to that of the allowed line is given, for weak microwave field intensities where the perturbation treatment of the problem is applicable[1,3,9] and for transitions of the type $l \to l \pm 2$, by

$$S_\pm{}^{(\pi)}(\theta) = \frac{e^2 a_0{}^2}{5\hbar^2} \frac{\langle E_\omega{}^2 \rangle}{(\Delta \pm \omega)^2} \times R_{ll'}(\sin^2\theta + \tfrac{3}{4}\cos^2\theta) \tag{1}$$

if the polarization of the laser beam is parallel to the direction of polarization of the microwaves, and by

$$S_\pm{}^{(\sigma)}(\theta) = \frac{3}{4} \frac{e^2 a_0{}^2}{5\hbar^2} \frac{\langle E_\omega{}^2 \rangle}{(\Delta \pm \omega)^2} R_{ll'} \tag{2}$$

if the laser beam is polarized perpendicular to the microwaves. These formulas are readily obtained from Ref. 3. θ is the angle between the incident laser beam and the direction of the microwave field E_ω, and Δ the splitting of the levels j and f measured in angular-frequency units. The meaning of the other symbols is identical to that in other publications.[1-3] The minus sign in the above formulas corresponds to the two-photon absorption process, the plus sign to stimulated Raman scattering, i.e., the absorption of an optical photon and the simultaneous emission of a microwave photon.

The experimental arrangement is essentially the same as that described in Ref. 7. The plasma source consists basically of a hollow cathode discharge operated at a filling pressure of 1.5 Torr helium with a discharge current of 12 mA. Superimposed on this dc plasma is a 1.5-kV pulse from a 4-μF capacitor which produces a plasma of much stronger line emission in the region between the electrodes. This plasma lasts for about 1 μsec; the electron density is of the order of 10^{13} cm^{-3}, the density of the neutrals $N_{\mathrm{He}} \simeq 5\times10^{16}$ cm^{-3}. As discussed previously[7] the assignment of an electron temperature is rather questionable.

The tunable dye laser for the scattering experiment is transversely pumped with the frequency-doubled beam from a Q-switched ruby laser and uses a 2.5×10^{-3} M solution of 7-diethylamino-4-methylcoumarin in ethanol. The laser cavity is formed by an echelle grating and a 50% reflecting dielectric output mirror and contains also a Fabry-Perot etalon. The spectral width thus obtained is 0.1 Å at output powers of the order of 10 kW. The echelle grating is rotated for coarse tuning of the laser wavelength, the Fabry-Perot etalon for fine tuning; it can be rotated to shift the laser wavelength by about 1 Å without affecting strongly the laser power. The resettability of the wavelength of the laser is better than 0.1 Å. The laser beam is polarized (90% of the intensity is in the horizontal plane, 10% in the vertical plane). The light scattered by the plasma is observed at 90° to the incident beam, and spectrally analyzed using a 1-m monochromator equipped with a photomultiplier. In addition, perpendicular to the incident beam and in the direction of observation, we also focus simultaneously microwave radiation of power of about 5 kW into the plasma. The microwaves have a frequency of 74 GHz and are generated by a magnetron model Amperex DX 164; the pulse duration is 1 μsec, and they are focused to a spot size approximately 1 cm in diameter using a spot-focusing conical horn lens.

The microwaves can alter of course our plasma conditions; any such change is, on the other hand, immaterial for the effects we want to observe: The laser pulse has a time duration of 11 nsec, and its effects can thus easily be observed on the quasistationary line emission. One can see the influence of the microwaves on the plasma by observing the line emission from the plasma. We time the laser pulse to occur about 200 nsec after the beginning of the microwave pulse, and at this time the microwave pulse has increased the line emission by less than 5%.

As the first experiment we tried the standard emission technique[1-3] and looked for the satellites due to the microwave field in the vicinity of the He I 4471-Å line. Using a time resolution of about 16 nsec limited by the Tektronix oscilloscope 551, the satellites were not sufficiently strong to be discernable from the noise.

In the second experiment we used the dye laser, tuned it to the He I 4471-Å line and scanned again the spectrum in the vicinity of this line. Because of the absorption of the laser light the 4471-Å line showed an enhancement of about a factor of 14 compared to the quasistationary line emission. Since the upper level of this line (4^3D) and the upper level of the forbidden transition (4^3F) are collisionally coupled,[7] the same enhancement can be seen on the satellites, which now become observable with high time resolution. The spectrum thus obtained is shown in Fig. 2: The intensity of the near satellite in emission relative to the allowed line is $S_-^{(e)} \simeq 3.9\times10^{-3}$, whereas the far satellite is only barely discernable. By attenuating the laser beam, we investigated the enhancement as a function of the laser power focused into the plasma. As the applied power was increased to the full laser power, the transition became saturated because the absorption was increasingly canceled by stimulated emission. This technique of pumping the allowed line is important for plasma diagnostic reasons and improves the standard emission technique[1-3]: It not only extends the sensitivity and allows thus to measure *local* electric fields by scanning the position of the dye laser path through the plasma. The increase of the sensitivity is, of course, limited by the saturation effect mentioned above.

The most powerful improvement with respect to

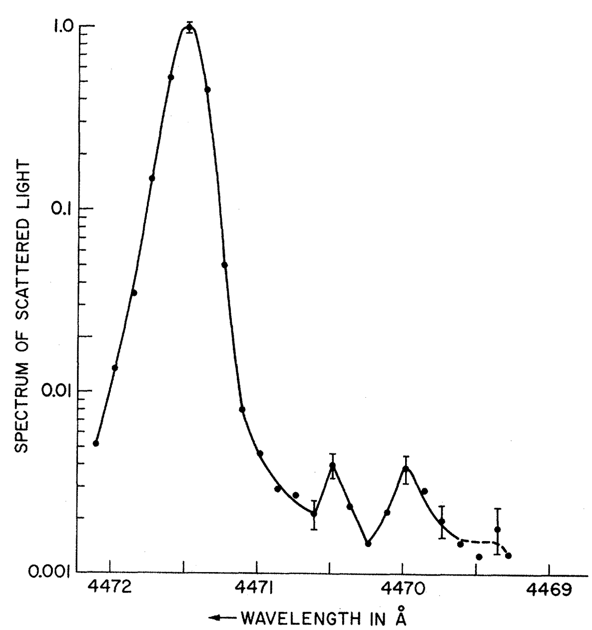

FIG. 2. Spectrum in the vicinity of the He I 4471-Å line due to absorption of laser light at the center of this line (intensity in arbitrary units).

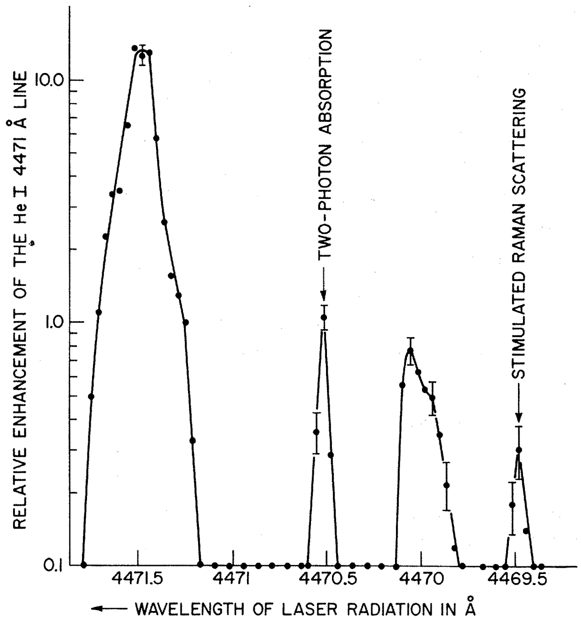

FIG. 3. Relative enhancement of the intensity of the He I 4471-Å line as a function of the wavelength of the incident laser radiation.

the standard emission technique, however, is demonstrated with the third experiment. Here we observed the He I 4471-Å line fixed and scanned the wavelength of the dye laser in its vicinity keeping the laser power constant. The results are shown in Fig. 3. We see the measured enhancement of the 4471-Å line due to the incident laser radiation as function of the wavelength of the laser beam. The minimum measurable enhancement of the 4471-Å line was 10% as a result of statistical fluctuations. The far satellite corresponding to stimulated Raman scattering and the near one corresponding to the two-photon absorption process are clearly observable and their "widths" are determined by the spectral width of the laser beam. The line corresponding to absorption at the He I 4471-Å one itself appears broad and has a flat top: This is due to the saturation of the absorption mentioned earlier. The "satellite intensities" $S_\pm$ in absorption according to Eqs. (1) and (2) are determined by the ratio of the laser power required to produce equal enhancement of the allowed line when the allowed transition is pumped to that required when the satellite transition is pumped. We thus obtained $S_-^{(\sigma)} \simeq 3.1\times 10^{-3}$ and $S_+^{(\sigma)} \simeq 9.3\times 10^{-4}$, in agreement with the emission values from Fig. 2. (In our case we practically see the absorption of the σ component only and have $\theta = 0°$.) The average microwave field thus measured is $\langle E^2\rangle^{1/2} \simeq 1.3$ kV/cm.

It is interesting to note that at the position of the forbidden line, we observe a relatively broad line, which is considerably broader than the satellites. Its width, the intensity of this line relative to the allowed line, as well as the width of the allowed line show[11] that this line cannot be the forbidden line due to particle-produced fields in the plasma, but must be produced by wave fields in the plasma, whose spectrum is not yet resolved because of the spectral width of the laser.

If we compare the three experiments—no satellites observed using the standard emission technique, satellites barely observable when pumping the allowed line (Fig. 2), and now the results of Fig. 3—the potentialities of the last measurements become obvious and the advantages of this technique can be summarized as follows: (1) One can obtain a *local* measurement of the electric fields in the plasma. (2) The sensitivity of this method can be greatly increased since one only has to increase the laser power (which should, of course, not change the plasma conditions). (3) This method discriminates against impurity lines. (4) The spectral resoltuion is limited only by the spectral width of the laser beam and the width of the forbidden line due to the particle-produced fields and Doppler broadening; the instrument function of the monochromator does not enter.

The stimulated Raman scattering observed here has recently also been suggested by Burgess, Richards, and Mahon[12] as a possibility of optically generating plasma waves in a plasma if higher laser powers become available.

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Scaling Equation of State for Thermodynamic Systems Having a Tricritical Point*

Peter J. Kortman†
Applied Mathematics Department, Brookhaven National Laboratory, Upton, New York 11973

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A phenomenological equation of state is presented which describes a thermodynamic system having a tricritical point. The equation is an asymptotic one—in the sense of ordinary critical-point theory—which obeys certain scaling relations and is consistent with Griffiths-Wheeler theory. It relates all tricritical exponents to the underlying second-order exponents by introducing a new exponent which determines the geometry of certain first-order phase lines at the tricritical point. The predictions are in reasonable agreement with experimental and numerical results.

The successful use of homogeneous functions (scaling functions) to represent thermodynamic systems near critical points is now well established.[1-3] Recent generalizations of the homogeneity postulate have been used to discuss systems having a tricritical point,[4,6] though unlike simple critical-point scaling theory, there is (to the best of our knowledge) no explicit equation of state to interrelate recent experimental results for critical exponents and to predict actual values for the singular part of thermodynamic functions. It is the purpose of this paper to introduce a phenomenological tricritical equation of state which can give nonclassical tricritical exponents that are in reasonable agreement with experimental and numerical results.[7] Apart from its potential use in predicting the behavior of measurable quantities and in fitting experimental data, our equation is important because it shows how the Griffiths-Wheeler theory, tricritical scaling theory, and the anticipated analytic properties of the tricritical free energy (real analyticity off phase surfaces) can be combined with the physical idea of competitive interactions to describe nonclassical tricritical behavior. Our equation of state is an extension of the Josephson-Schofield scaling formulation[8] and has been obtained by generalizing a Schofield representation for the tricritical mean-field model.[9] However, unlike the mean field, our equations predict nonclassical exponents except on the tricritical wings where the exponents do appear to be classical.

As an example of a tricritical system, we consider the metamagnet $FeCl_2$.[10] A plausible phase diagram for this spin-1, Ising-like antiferromagnet is shown in Fig. 1. The variables H, T, and H_S represent internal magnetic field, temperature, and staggered magnetic field, respectively. It is convenient to parametrize the ($H_S = 0$) second-order line and its analytic continuation past the tricritical point (TCP) by the field H. This curve, labeled AA' in Fig. 2, is denoted by $T_c(H)$ which we assume to be an analytic function. Close to the line $T_c(H)$ for $H < H_t$ we expect that simple Schofield theory[8] will accurately describe the critical region provided the coefficients in the Schofield equations are made suitably H and T dependent.[11] We assume that near the TCP the important variation in these coefficients depends on the function λ defined by $\lambda(T,H) \equiv a(H-H_t) + b(T-T_t)$, where a and b are constants and T_t is the tricritical temperature. The line BB' in

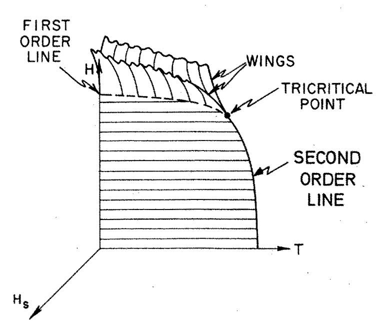

FIG. 1. Plausible phase diagram for the metamagnet $FeCl_2$. The lined surfaces are first-order surfaces.