Measurement of Fokker-Planck Diffusion with Laser-Induced Fluorescence

J. J. Curry, F. Skiff, and M. Sarfaty
Institute for Plasma Research, University of Maryland, College Park, Maryland 20742-3511

T. N. Good
Department of Physics, Gettysburg College, Gettysburg, Pennsylvania 17325
(Received 1 November 1993)

Laser-induced fluorescence in plasmas is shown to be strongly influenced by velocity-space diffusion, under the proper conditions. The induced fluorescence is modeled by a set of coupled rate equations that include the Fokker-Planck operator for each state density, and the results are compared with data from a gas-discharge plasma in which the ArII $(3d')^2\,G_{9/2}$ metastable state is optically pumped. The Fokker-Planck diffusion coefficient D is determined and found to be in agreement with theory.

PACS numbers: 52.25.Fi, 52.70.Kz

Transport phenomena in plasmas are a topic of fundamental importance, both for obtaining an understanding of the basic physical mechanisms that move an ionized gas toward and also maintain equilibrium as well as for the development of advanced applications [1–3]. One aspect of the transport problem is the diffusion of a test particle through a medium already in equilibrium. Test-particle velocity-space, or Fokker-Planck, diffusion has recently been measured [4] through a two-step process called "optical tagging" [5] in which a narrow-band laser spectroscopically "tags" a given velocity class of ions. A second narrow-band laser is subsequently used to view, by means of laser-induced fluorescence (LIF), the "tagged" ions as they diffuse. In this Letter we will show that information about diffusion can also be obtained with just one laser by correctly analyzing the resultant time-dependent fluorescence. We present the first measurements of velocity-space Coulomb diffusion in a gas-discharge plasma.

The time dependence of sub-Doppler LIF can be influenced by many factors including laser intensity and spectral width, collisional excitation rates, Zeeman splitting, etc. In systems where optical pumping occurs, diffusion also needs to be considered. If the Fokker-Planck coefficient D and Doppler width of the optically pumped region σ are such that D/σ^2 is at least comparable with the optically stimulated transition rate, then diffusion is significant and is therefore evident in the time evolution of the induced fluorescence. We have observed such evidence in a magnetized argon (II) plasma. A system of rate equations governing the time-dependent populations of the appropriate ionic states, and therefore the time-dependent LIF signal, has been developed. The primary difference between this system and previous treatment [6,7] is the inclusion of a Fokker-Planck operator. By using these equations to simulate the expected LIF signal, quantitative information can be obtained about velocity-space diffusion in the plasma [6,7].

The experiment was performed in an ArII plasma, immersed in a solenoidal magnetic field of approximately 2500 G. The 10 cm diam plasma is continuously produced by nonresonant radio-frequency excitation. A schematic of the apparatus is shown in Fig. 1. The laser diagnostic consists of a Coherent 599-21 single-mode continuous-wave dye laser in conjunction with an external acousto-optic modulator. The output of the dye laser, before modulation, has a bandwidth on the order of 1 MHz, and a center frequency that is electronically tunable. The modulator was used to produce 60 μs laser pulses separated by several milliseconds. The laser beam is then directed through the plasma, parallel to the magnetic field. Fluorescence induced by this beam is collected along the radial direction at the vacuum port shown. A mirror on

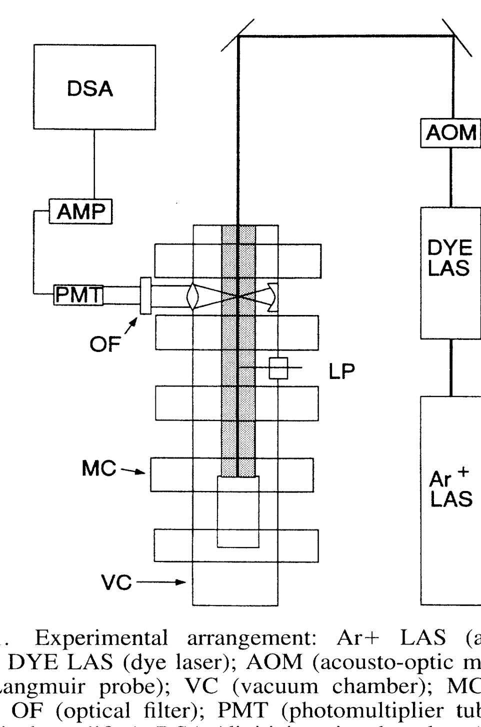

FIG. 1. Experimental arrangement: Ar+ LAS (argon ion laser); DYE LAS (dye laser); AOM (acousto-optic modulator); LP (Langmuir probe); VC (vacuum chamber); MC (magnet coils); OF (optical filter); PMT (photomultiplier tube); AMP (electrical amplifier); DSA (digitizing signal analyzer).

one side of the plasma and a lens on the other collect and collimate the light before it passes through an optical bandpass filter and then strikes a photomultiplier tube. The electrical signal is amplified and sent to a Tektronix Digitizing Signal Analyzer. Noise, resulting primarily from fluctuations in the background light from the plasma, is eliminated by averaging wave forms at each laser frequency (ion velocity). The other available diagnostic is a Langmuir probe which is used to measure electron temperature T_e and ion density n_{ion}.

The LIF process is depicted in the Grotrian diagram of Fig. 2. We chose to pump the ArII excited metastable state $(3d')^2 G_{9/2}$ because we found it to be well populated and accordingly the laser was tuned to 611 nm. The upper state of this transition, $(4p')^2 F_{7/2}$, spontaneously decays primarily to the $(4s')^2 D_{5/2}$ state, emitting fluorescence at 461 nm [8]. The dynamics of these state densities, and therefore the emitted fluorescence, in the presence of a resonant optical field can be modeled by a set of coupled first-order ordinary differential equations. However, when the ensemble of ions is characterized by a distribution of velocities, the equations become velocity dependent. The optically pumped ions create a "test-ion" distribution that has an average velocity and width determined by the laser. Furthermore, these test ions interact with the ions of the background distribution through the local electric field. We have used the following two coupled partial differential equations to describe the populations of the metastable level (1) and upper level (2):

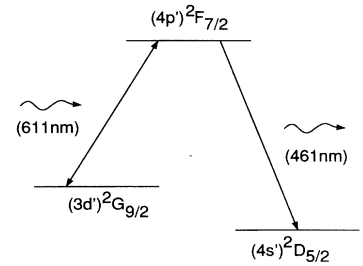

FIG. 2. The dye laser, at 611 nm, induces fluorescence at 461 nm by pumping the excited ArII metastable state $(3d'^2)G_{9/2}$.

$$\frac{dn_1}{dt} = -\left[B_{12}\frac{I}{c}g(v_z) + r_1 + r_{12}\right]n_1 + \left[B_{21}\frac{I}{c}g(v_z) + A_{21} + r_{21}\right]n_2 + \left.\frac{\partial n_1}{\partial t}\right|_{\mathrm{FP}} + \gamma_1, \quad (1)$$

$$\frac{dn_2}{dt} = \left[B_{12}\frac{I}{c}g(v_z) + r_{12}\right]n_1 - \left[B_{21}\frac{I}{c}g(v_z) + A_{\mathrm{total}} + r_2 + r_{21}\right]n_2 + \left.\frac{\partial n_2}{\partial t}\right|_{\mathrm{FP}} + \gamma_2, \quad (2)$$

where

$$n_i = n_i(\vec{x}, v_z, t) = \int f_i(\vec{x}, \vec{v}, t)\, dv_x\, dv_y \quad (3)$$

and $f_i(\vec{x}, \vec{v}, t)$ is the phase-space distribution of ions in the ith level. A and B are the Einstein spontaneous and stimulated rate coefficients, respectively, I is the laser intensity, and v_z is the component of velocity parallel to the beam. Note that the velocity dependence has been reduced to one dimension because the laser is velocity selective only along its direction of propagation. In this case the laser spectral width of 1 MHz is considerably smaller than the natural linewidth ($\Delta\nu = 18.9$ MHz) of the 611 nm transition, so

$$g(v) = \frac{\lambda\sigma}{2\pi[(v - v_0)^2 + (\frac{\sigma}{2})^2]}, \quad (4)$$

$$\sigma = \lambda\Delta\nu = \lambda A_{\mathrm{total}}/2\pi, \quad (5)$$

is the natural Lorentzian line shape converted to velocity dependence through the relation $v - v_0 = \lambda(\nu - \nu_0)$. It should be noted that Eqs. (1) and (2), with the definitions given above, provide for optical saturation effects [9,10]. r_1 and r_2 represent loss rates of the corresponding states (not including transitions between the two), possibly resulting from collisions or flow, and r_{12}, r_{21} are the collisionally induced transition rates between states 1 and 2. γ_1 and γ_2 are creation rates that depend on the background plasma. Finally, we must account for the interaction of test ions with the background distribution. This is done through the Fokker-Planck diffusion operator [11],

$$\left.\frac{\partial n(\vec{x}, v_z, t)}{\partial t}\right|_{\mathrm{FP}} = \frac{\partial}{\partial v_z}\left[-Cn(\vec{x}, v_z, t) + D\frac{\partial}{\partial v_z}n(\vec{x}, v_z, t)\right], \quad (6)$$

$$C \equiv \left\langle\frac{\delta v_z}{\delta t}\right\rangle - \frac{\partial}{\partial v_z}\left\langle\frac{\delta v_z \delta v_z}{\delta t}\right\rangle, \quad (7)$$

$$D \equiv \left\langle\frac{\delta v_z \delta v_z}{\delta t}\right\rangle. \quad (8)$$

This operator is based on the picture that each test ion continually receives many small "kicks," δv_z, from the local electric field (arising from fluctuations in the background ion distribution). These kicks may be the result of large amplitude waves (anomalous diffusion) or thermal fluctuations. The coefficients C and D link macroscopic changes of the test-ion distribution to kicks occurring on the microscopic level. When the background distribution is Maxwellian, C and D are not independent but are related as follows:

$$-C = (v/v_{\mathrm{th}}^2)D. \quad (9)$$

These kicks, then, can often be described by a single function.

The effect of the Fokker-Planck term on the solution of the above rate equations is qualitatively different than that of the other terms. Unlike constant rate coefficients, its magnitude is a strong function of the shape of the test-ion distribution, particularly through the second derivative. In order that diffusion play a significant role, the following relationships must hold:

$$r_1 < B_{12} I g(v_0)/c < D/\sigma^2 . \qquad (10)$$

The first inequality requires that some optical pumping occur, i.e., a test-ion distribution must be created and these ions must have a chance to diffuse out of resonance with the laser. The second inequality states that the laser spectral intensity not be so strong as to completely dominate. This can happen if the wings of the laser spectrum contain enough power that the test-ion distribution spreads faster by optical pumping than by diffusion.

Table I lists the measured characteristics of the plasma. From these we can calculate the parameters needed for the model. The decay rates A_{total} and A_{12} can be found in the literature [8] and used to determine B_{ij}. The creation rates γ_1 and γ_2 play no role in the dynamics of the solution, only in determining equilibrium populations. Also, r_2 is not critical since it is typically negligible compared to A_{total}. We have estimated r_{ij} by considering electron-ion inelastic collisions. The appropriate rate coefficient is given by McWhirter [12]:

$$r_{12} = \frac{6.5 \times 10^{-4}}{\Delta E T_e^{1/2}} n_{\text{electron}} f \exp\left(-\frac{\Delta E}{T_e}\right), \qquad (11)$$

where f is the oscillator strength and ΔE (in eV) is the change in energy of the transition. Because we have a singly ionized plasma, quasineutrality requires $n_{\text{electron}} = n_{\text{ion}}$ resulting in $r_{12} \approx 2 \times 10^4\ \text{s}^{-1}$. ($r_{21}$ equals r_{12} without the exponential factor. However, it is still insignificant compared to A_{21}.) It must be realized that this formula provides only an order of magnitude estimate of the desired rate [13], and we have found that the value $r_{12} = 1 \times 10^4\ \text{s}^{-1}$ provides a better fit to the data. Estimating r_1 is a bit more difficult. Electron-ion collisions, charge exchange, as well as cross-field flow must be considered. We are not certain which effect is dominant in determining the metastable lifetime in the laser beam. Fortunately, the effect of r_1 is distinguishable from that of diffusion, so it is not necessary to know r_1 *a priori*. We have found that $r_1 = 3 \times 10^4\ \text{s}^{-1}$ is appropriate.

TABLE I. Plasma characteristics and their uncertainties. All of these values were determined independently of diffusion measurements.

Parameter	Value	Uncertainty
Ion density	$3 \times 10^9\ \text{cm}^{-3}$	50%
Ion temperature	0.05 eV	10%
Electron temperature	3 eV	15%
$\langle \delta n \rangle / n$	0.03	...
Parallel ion flow	1.5×10^4 cm/s	...
Laser intensity	10 mW/cm^2	50%
Magnetic field	2450 G	2%

With the model we have developed we can show that under the appropriate conditions (i) Fokker-Planck diffusion is evident in the time evolution of laser-induced fluorescence and its manifestation in LIF is distinct from that of other terms, and (ii) quantitative information about D can be obtained. In Fig. 3 are shown three normalized traces: the observed fluorescence and two results from the model. The dashed trace was produced by eliminating the Fokker-Planck operator from the model. With the given parameters (even allowing for uncertainties), it does not predict the fluorescence that we observe. The solid trace, however, because of its inclusion of the Fokker-Planck term, is capable of fitting the data very well. By comparing these two traces it can be seen that diffusion is significant and also how it is manifested in LIF. First, the asymptotic level is higher, as if the metastable state lifetime had been reduced. Second, and perhaps more importantly, diffusion begins to act very quickly on the leading edge of pulse, as soon as the test-ion distribution is created. Its effect is to reduce the apparent optical pumping. With diffusion, fluorescence decay is much slower, consistent with considerably lower laser power. However, the laser spectral intensity can be measured and this allows a quantitative measurement of D by using the model to fit the data. The result is that we find $D(v = 0) = (2.2 \pm 0.2) \times 10^{13}\ \text{cm}^2\,\text{s}^{-3}$. To compare this with the theoretical prediction appropriate for a "quiet" plasma [4,14],

$$D(v) = -\frac{\pi n e^4}{m_i^{3/2} T_i^{1/2}} \ln\left[\frac{3}{2}\left(\frac{T_i^3}{\pi n}\right)^{1/2} \frac{1}{e^3}\right] \frac{1}{x} \frac{\partial}{\partial x}\left(\frac{\text{erf}(x)}{x}\right), \qquad (12)$$

$$x = v/\sqrt{2} v_t , \qquad (13)$$

we use T_i and n_{ion} as determined by separate LIF and Langmuir probe measurements. The result is $D(v = 0) = (3 \pm 1.5) \times 10^{13}\ \text{cm}^2\,\text{s}^{-3}$, the uncertainty due principally to the ion density measurement with the Langmuir probe.

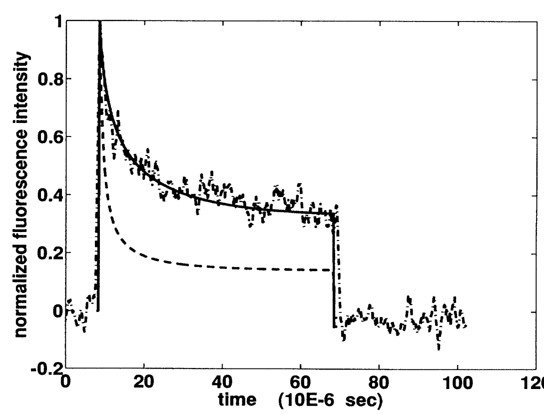

FIG. 3. Our model shows the importance of including diffusion in the rate equations. Including diffusion significantly alters the dynamics of optical pumping (· — · — · LIF data: — model with diffusion; - - - model without diffusion).

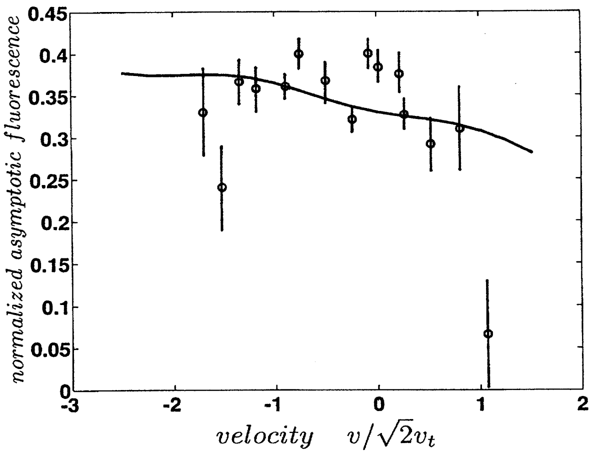

FIG. 4. The model predicts that the magnitude of Zeeman splitting in this experiment reduces velocity variation of the induced fluorescence (∘ experimental data; — model prediction).

The normalized asymptotic fluorescence shows little variation with velocity. To demonstrate this graphically, we have plotted, in Fig. 4, the normalized asymptotic level (a parameter that is sensitive to diffusion) versus velocity for both the data and model predictions. The lack of variation, which is also predicted by the model, is to be expected given the degree of Zeeman splitting and the low ion temperature. Consequently, we are not able to determine the velocity dependence of D in this case. This is a limitation imposed only by the experimental situation and not by the method.

It is also interesting to note how the measured diffusion scales with density, as this parameter is prominent in the theoretical result for the quiet plasma case. In a separate experiment, diffusion was found to be linear with density, as shown in Fig. 5, over the range covered. However, the absolute level of diffusion is about a factor of 4 above that predicted by Eq. (12). This is not surprising because the data of Fig. 5 were obtained from a plasma with a much higher level of density fluctuations than the data in previous figures. Equation (12), on the other hand, was derived in the limit of minimum thermal fluctuations, a condition usually approached only in Q-machine plasmas where measurements have shown good agreement with Eq. (12).

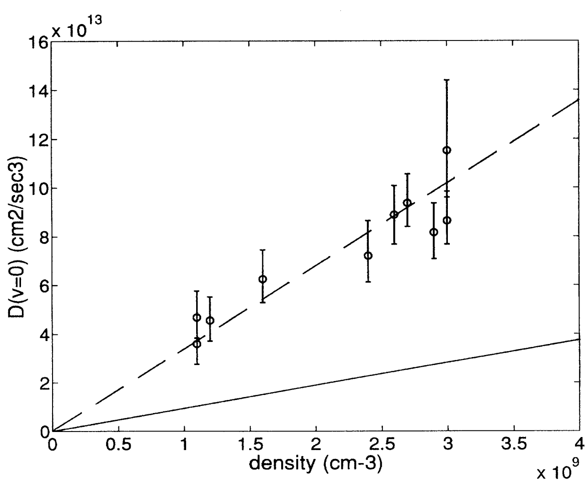

FIG. 5. The variation of the measured diffusion coefficient with density in a noisy plasma is linear (dashed line). The theoretical "quiet" plasma results [Eq. (13)] are shown for comparison (solid line).

In this Letter, we have shown that, under proper conditions, the effects of test-particle diffusion are apparent in the time evolution of laser-induced fluorescence. With the use of a rate-equation model, the magnitude of the diffusion coefficient D has been measured and found to be consistent with theoretical predictions for a quiet plasma. In an experiment where the velocity resolution of LIF is not severely limited by Zeeman splitting or other factors, the velocity dependence of D should be obtainable with this technique.

This work was supported by National Science Foundation Grant No. PHY-9224548.

[1] R.T. Farouki, S. Hamaguchi, and M. Dalvie, Phys. Rev. A **44**, 2664 (1991).
[2] N. Sadeghi, J. Appl. Phys. **70**, 2552 (1991).
[3] G.S. Selwyn, J. Vac. Sci. Technol. B **9**, 3487 (1991).
[4] J. Bowles, R. McWilliams, and N. Rynn, Phys. Rev. Lett. **68**, 1144 (1992).
[5] R.A. Stern, D.N. Hill, and N. Rynn, Phys. Lett. **93A**, 127 (1983).
[6] R.A. Stern, Phys. Fluids **21**, 1287 (1978).
[7] D.N. Hill, Ph.D. dissertation, University of California, Irvine, 1983. (Hill's thesis treats time dependence.)
[8] V. Vujnovic and W.L. Wiese, J. Phys. Chem. Ref. Data **21**, 919 (1992).
[9] M.J. Goeckner and J. Goree, J. Vac. Sci. Technol. A **7**, 977 (1989).
[10] M.J. Goeckner, J. Goree, and T.E. Sheridan, Rev. Sci. Instrum. **64**, 996 (1993).
[11] N.A. Krall and A.W. Trivelpiece, *Principles of Plasma Physics* (San Francisco Press, San Francisco, 1986), p. 295; R. Balescu, *Transport Processes in Plasmas* (Elsevier Science Publishers, New York, 1988), Vol. 2, p. 450.
[12] R.W.P. McWhirter, in *Plasma Diagnostics,* edited by R.H. Huddlestone and S.L. Leonard (Academic, New York, 1965), p. 206.
[13] H.R. Griem, in *Radiative Processes in Discharge Plasmas,* edited by J.M. Proud and L.H. Luessen, NATO ASI Ser. B, Vol. 149 (Plenum, New York, 1986), p. 15.
[14] N. Rostoker and M.N. Rosenbluth, Phys. Fluids **3**, 1 (1960); W.B. Thompson and J. Hubbard, Rev. Mod. Phys. **32**, 714 (1960).