[10]See, for example, J. O. Hirschfelder, C. F. Curtis, and R. B. Bird, *Molecular Theory of Gases and Liquids* (Wiley, New York, 1966).

[11]At room temperature for He the estimate can be performed classically as quantum effects are very small and can be neglected for our purposes.

[12]For the L-J potential parameters see Hirschfelder, Curtis, and Bird, Ref. 10.

[13]F. Barocchi, M. Neri, and M. Zoppi, Mol. Phys. 34, 1391 (1977).

[14]The expressions for the moments is given in H. B. Levine, J. Chem. Phys. 56, 2455 (1972).

Theory of Atomic Motion in a Resonant Electromagnetic Wave

R. J. Cook

University of California, Lawrence Livermore Laboratory, Livermore, California 94550

(Received 6 November 1978)

A new theory of atomic motion in a resonant standing or traveling electromagnetic wave is presented. It is shown that, when effects of spontaneous emission are negligible, the motion of a two-level atom in the resonant radiation is determined by two noninterfering wave functions, each of which satisfied a time-dependent Schrödinger equation with time-independent potential energy. An experiment is proposed to test the theory.

There has been renewed interest in recent years in the theory of atomic motion in resonant and near-resonant electromagnetic waves. This interest was initiated by the development of high-power tunable lasers, and interest has continued because of possible application of the theory to problems of laser isotope separation,[1-4] atomic trapping and cooling,[4-7] neutral-atom acceleration,[8-9] and atomic-beam–deflection spectroscopy.[10-12]

An atom illuminated by resonant radiation experiences at least two types of radiation force: a force associated with spontaneous emission, and a force due to interaction of the induced atomic dipole moment with the amplitude gradient of the applied field. The radiation pressure associated with spontaneous emission has been extensively investigated both theoretically[4,6] and experimentally,[1,12,13] and will not be discussed in this Letter. A theory of the induced-dipole force has been developed by Ashkin for the case where the atomic response may be described by a polarizability,[6] and a certain off-resonance focusing effect associated with this force has recently been detected experimentally.[14] A description of the atomic response in terms of a polarizability also involves spontaneous emission to the extent that it is spontaneous decay that causes atomic relaxation to the near–steady-state condition described by a polarizability. If the atom-field interaction is brief (less than a natural lifetime), a theory based on the steady-state polarizability is no longer appropriate, and, in general, Schrödinger's equation must be solved to determine the atomic response. In this case, effects of spontaneous emission are negligible.

The purpose of this Letter is (1) to present a new theory of atomic motion in a resonant electromagnetic wave, applicable when effects of spontaneous emission are negligible, and (2) to propose an experimental test of the new theory.

The Hamiltonian for an atom in a classically prescribed electromagnetic field, in the dipole approximation, takes the form

$$H = P^2/2M + H_0 - \vec{\mu}\cdot\vec{E}(\vec{R}, t), \tag{1}$$

where $P^2/2M$ is the kinetic energy associated with the center-of-mass momentum $\vec{P}$, H_0 is the Hamiltonian for the internal motion of the atom, $\vec{\mu}$ is the dipole-moment operator, and $\vec{E}(\vec{R}, t)$ is the electric field evaluated at the center-of-mass position $\vec{R}$. Consider first the motion of a two-level atom with energy levels E_1 and E_2 in a monochromatic standing wave $\vec{E}(\vec{x}, t) = \hat{\epsilon}\mathcal{E}(\vec{x})\cos\omega t$. Here the amplitude $\mathcal{E}(\vec{x})$ will be a solution of the time-independent wave equation $\nabla^2\mathcal{E} + (\omega/c)^2\mathcal{E} = 0$, but is otherwise arbitrary. Let $\psi_1(\vec{x})$ and $\psi_2(\vec{x})$ be the amplitudes for the atom to be located at position $\vec{x}$ and occupy energy levels E_1 and E_2, respectively. Then it follows from Eq. (1) that the Schrödinger equation for the two-component wave function is

$$\begin{aligned} i\hbar\frac{\partial\psi_1}{\partial t} &= -\frac{\hbar^2}{2M}\nabla^2\psi_1 + E_1\psi_1 - \mu\mathcal{E}(\vec{x})\cos\omega t\,\psi_2, \\ i\hbar\frac{\partial\psi_2}{\partial t} &= -\frac{\hbar^2}{2M}\nabla^2\psi_2 + E_2\psi_2 - \mu\mathcal{E}(\vec{x})\cos\omega t\,\psi_1, \end{aligned} \tag{2}$$

where $\mu = \langle 1 | \vec{\mu}\cdot\hat{\epsilon} | 2\rangle$ is the transition dipole moment. In the case of exact resonance, $\omega = (E_2$

$-E_1)/\hbar$, the substitutions $\psi_1 = C_1 \exp(-iE_1 t/\hbar)$ and $\psi_2 = C_2 \exp(-iE_2 t/\hbar)$, and a rotating-wave approximation, i.e., neglect of inessential terms that oscillate at twice the optical frequency, put Eqs. (2) in the form

$$i\hbar \frac{\partial C_1}{\partial t} = -\frac{\hbar^2}{2M}\nabla^2 C_1 - \tfrac{1}{2}\mu \mathcal{E}(\vec{x}) C_2,$$
$$i\hbar \frac{\partial C_2}{\partial t} = -\frac{\hbar^2}{2M}\nabla^2 C_2 - \tfrac{1}{2}\mu \mathcal{E}(\vec{x}) C_1. \quad (3)$$

Equations (3) are decoupled by the unitary transformation

$$u_+ = 2^{-1/2}(C_1 - C_2),$$
$$u_- = 2^{-1/2}(C_1 + C_2). \quad (4)$$

The equations of motion for $u_\pm(\vec{x})$ are

$$i\hbar \frac{\partial u_\pm}{\partial t} = -\frac{\hbar^2}{2M}\nabla^2 u_\pm \pm \tfrac{1}{2}\mu \mathcal{E}(\vec{x}) u_\pm, \quad (5)$$

and the probability density for the position of the atom, $P(\vec{x}) = |\psi_1(\vec{x})|^2 + |\psi_2(\vec{x})|^2$, becomes

$$P(\vec{x}) = |u_+(\vec{x})|^2 + |u_-(\vec{x})|^2. \quad (6)$$

Equations (5) state that the wave functions $u_\pm$ propagate independently of each other, and Eq. (6) shows that there is no spatial interference between these waves. An atom initially in the ground state ($C_2 = 0$) has equal probability to be in one or the other of the waves $u_\pm$, and Eqs. (5) imply that these probabilities are time independent. The waves $u_\pm$ each satisfy a simple time-dependent Schrödinger equation, but with potential energies $V_\pm(\vec{x}) = \pm\frac{1}{2}\mu\mathcal{E}(\vec{x})$ of opposite sign. Therefore the forces acting on atoms in the two waves are in opposite directions, and it follows that the amplitude gradient of the resonant radiation will split a narrow atomic beam into two components, in much the same way as the magnetic field gradient splits an atomic beam in the Stern-Gerlach experiment. Equations (5) and (6) are consistent with the theory of Kazantsev that predicts splitting of an atomic trajectory upon crossing a sharp boundary between the vacuum and resonant field.[15]

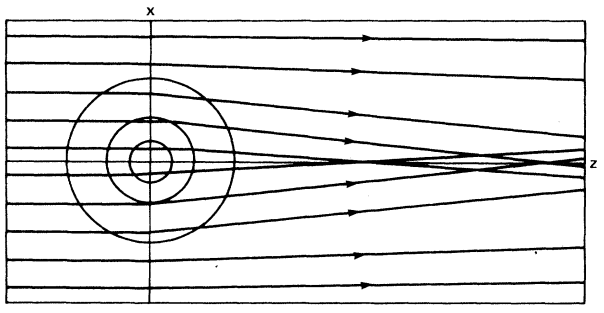

FIG. 1. Focusing of atomic trajectories associated with the wave function u_-.

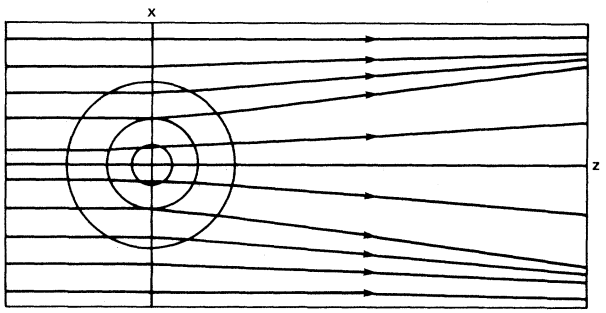

FIG. 2. Defocusing of atomic trajectories associated with the wave function u_+.

If N atomic levels take part in the resonant interaction, an analysis similar to the above shows that the motion of the atom is determined by N independent noninterfering wave functions u_n. The wave function u_n satisfies a time-dependent Schrödinger equation with potential energy $V_n(\vec{x}) = \frac{1}{2}\mu_n \mathcal{E}(\vec{x})$, where μ_n is the nth eigenvalue of the matrix of transition dipole moments connecting the N levels. The probability density for the position of the atom is $P(\vec{x}) = \sum_n |\mu_n(\vec{x})|^2$, and the probability that the atom occupies the wave u_n, for an atom initially in the ground state, is the absolute square of the first element of the nth eigenvector of the dipole-moment matrix.

When the resonant field is a traveling wave, $\vec{E}(\vec{x}, t) = \hat{\vec{\epsilon}}\mathcal{E}(\vec{x})\cos[\varphi(\vec{x}) - \omega t]$, with $|\nabla\varphi| \approx 2\pi/\lambda$, certain additional terms must be added to Eqs.

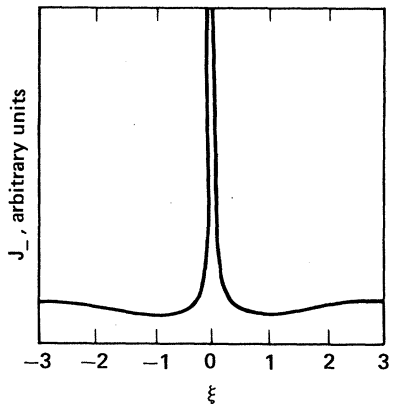

FIG. 3. Atomic flux J_- vs ξ in the focal plane $s = 1$.

(3). Under usual experimental conditions of well-collimated atomic and optical beams, these terms represent a simple Doppler shift. If the detuning $\Delta=\omega-\omega'$ $[\omega'=(E_2-E_1)/\hbar]$ is adjusted to cancel the Doppler shift, the above theory [Eqs. (3) and their consequences] is recovered to an excellent approximation.

The theory may be tested as follows. Consider a well-collimated atomic beam of ground-state two-level atoms that propagates in the z direction and intersects a collimated Gaussian optical beam $\mathcal{E}(\vec{x})=\mathcal{E}_0\exp[-(x^2+z^2)/\omega_0{}^2]$ propagating in the y direction. The resonant radiation acts as a cylindrical lens that tends to focus atoms in the wave u_- and defocus atoms in the wave u_+, as illustrated in Figs. 1 and 2, respectively. In crossing the Gaussian beam, the initial plane waves $u_\pm=\exp(ikz-i\nu t)$ acquire phase factors $\exp[\mp i\theta(x)]$, where $\theta(x)=\pi^{1/2}\omega_0\mu\mathcal{E}_0\exp(-x^2/\omega_0{}^2)/2\hbar v_z$, and subsequently propagate as free-particle wave functions. Let $\xi=x/\omega_0$, $s=z/f_0$, and $m=\pi^{1/2}\omega_0\mu\mathcal{E}_0/2\hbar v_z$, where v_z is the atomic velocity, $f_0=2\omega_0\epsilon/\pi^{1/2}\mu\mathcal{E}_0$ ($\epsilon=\frac{1}{2}Mv_z{}^2$) is the "focal length" of the Gaussian lens, and m is a dimensionless measure of the strength of the resonant field. Then the waves $u_\pm$ on the downstream side of the radiation (neglecting inessential phase factors) are given by

$$u_\pm(\xi,s)=(m/\pi s)^{1/2}\int_{-\infty}^{\infty}\exp\{im[s^{-1}(\xi-\xi_0)^2\mp\exp(-\xi_0{}^2)]\}d\xi_0. \tag{7}$$

Equation (7) is the result of a Fresnel approximation and is valid when atomic deflections are small. To the same approximation, the atomic flux associated with u_+ is $J_+(\xi,s)=v_z|u_+\xi,s)|^2$, the flux associated with u_- is $J_-(\xi,s)=v_z|u_-(\xi,s)|^2$, and the total flux is $J=J_++J_-$.

To be specific, consider a 1-W Gaussian laser beam focused to waist $\omega_0=50$ μm. The peak intensity is then $I_0=2.5\times10^4$ W/cm^2 and $\mathcal{E}_0=(8\pi I_0/c)^{1/2}$. Let the atomic beam issue from an oven at temperature $T=1000$ K, and select velocity component v_z equal to $\frac{1}{10}$th of the most probable thermal velocity. Then for "typical" atoms of mass $M=50u=8\times10^{-23}$ g and transition dipole moment $\mu=1$ D, the focal length of the radiation lens is $f_0\approx0.8$ cm, and $m=7.2\times10^3$.

The atomic fluxes $J_-(\xi)$ and $J_+(\xi)$ in the focal plane ($s=1$) are plotted in Figs. 3 and 4, respectively. The dominant feature of $J_-(\xi)$ is the sharp peak at $\xi=0$. This is the primary focal line of the radiation lens. The flux $J_+(\xi)$ shows two peaks, formed by atomic trajectories that are repelled by the resonant field. The total flux $J=J_++J_-$, Fig. 5, shows quite clearly the effects of both focusing and defocusing of atomic trajec-

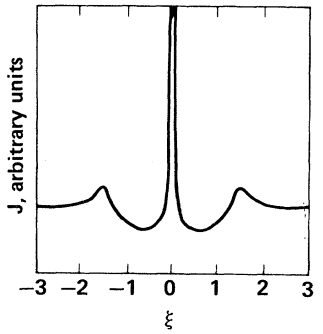

FIG. 5. Total atomic flux $J=J_++J_-$ vs ξ in the focal plane $s=1$.

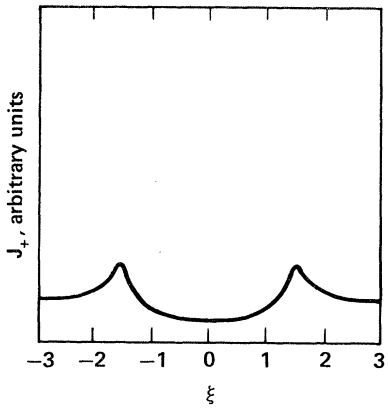

FIG. 4. Atomic flux J_+ vs ξ in the focal plane $s=1$.

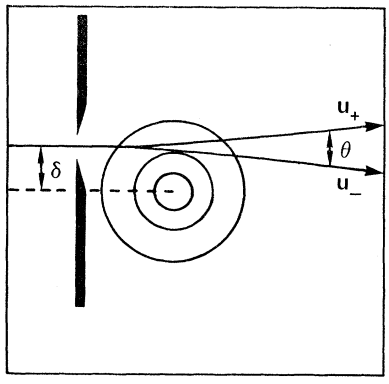

FIG. 6. Splitting of an atomic beam by the amplitude gradient of the resonant field.

tories. As s increases beyond $s=1$, the peaks of $J_+(\xi)$ become sharper and form two secondary focal lines in the plane $s \approx 2.3$ at $\xi \approx \pm 1.8$. These focal lines are a result of focusing by the wings of the negative Gaussian radiation lens. As s varies from 1 to 2.3, the central focal line of $J_-(\xi)$ spreads into a band of width $\Delta\xi \approx 1.0$. The focal lines of $J_+(\xi)$ lie well outside of this band. The structure of the atomic flux would be well resolved by a detector of resolution $\Delta x = 10$ μm.

A direct measurement of the splitting of an atomic beam by the amplitude gradient of the resonant field may be accomplished by placing a narrow slit immediately upstream of the interaction region, as illustrated in Fig. 6. The maximum splitting occurs when the center of the slit is off-axis by the amount $\delta = \omega_0/\sqrt{2}$, and, in the above example, has the value $\theta = 0.31°$. For a slit width of 20 μm, the divergence of each of the deflected components is less than 0.03°.

The present theory is valid when effects of spontaneous emission are negligible, i.e., when the interaction time is less than the natural lifetime of the atoms ($\Delta t = 2\omega_0/v_z < \tau_n$). In the above example, this condition obtains when the resonant frequency is less than 2.5×10^{15} Hz or $\lambda \gtrsim 0.7$ μm. This constraint may be relaxed somewhat by decreasing μ, M, and/or ω_0.

A measurement of the total atomic flux in the focal plane ($s=1$) showing the triple-peaked structure of $J(\xi)$, or a direct measurement of the splitting of an atomic beam by the amplitude gradient of the resonant field, would provide a convincing test of the present two-component theory.

The author wishes to acknowledge stimulating conversations with E. Teller, B. W. Shore, and A. F. Bernhardt during the course of this work. This work was performed under the auspices of the U. S. Department of Energy under Contract No. W-7405-ENG-48.

[1]A. F. Bernhardt, D. E. Duerre, J. R. Simpson, and L. L. Wood, Appl. Phys. Lett. 25, 617 (1974).
[2]Anthony F. Bernhardt, Appl. Phys. 34, 19 (1976).
[3]G. A. Delone, V. A. Grinchuk, A. P. Kazantsev, and G. I. Surdutovich, Opt. Commun. 25, 399 (1978).
[4]A. Ashkin, Phys. Rev. Lett. 24, 156 (1970).
[5]T. W. Hänsch and A. L. Schawlow, Opt. Commun. 13, 68 (1975).
[6]A. Ashkin, Phys. Rev. Lett. 40, 729 (1978).
[7]V. S. Letokhov, V. G. Minogin, and B. D. Pavlik, Opt. Commun. 19, 72 (1976).
[8]A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 66, 1599 (1974), and 63, 1628 (1973) [Sov. Phys. JETP 39, 784 (1974), and 36, 861 (1973)].
[9]A. Yu. Pusep, Zh. Eksp. Teor. Fiz. 70, 851 (1976) [Sov. Phys. JETP 43, 441 (1976)].
[10]A. F. Bernhardt, D. E. Duerre, J. R. Simpson, and L. L. Wood, Opt. Commun. 16, 166 (1976).
[11]A. Ashkin, Phys. Rev. Lett. 25, 1321 (1970).
[12]J. L. Picque and J. L. Vialle, Opt. Commun. 5, 402 (1972).
[13]R. Schieder, H. Walther, and L. Wöste, Opt. Commun. 5, 337 (1972).
[14]J. E. Bjorkolm, R. R. Freeman, A. Ashkin, and D. B. Pearson, Phys. Rev. Lett. 41, 1361 (1978).
[15]A. P. Kazantsev, Zh. Eksp. Teor. Fiz. 67, 1660 (1974) [Sov. Phys. JETP 40, 825 (1975)].

Effect of Vibrational and Rotational Exictation on Dissociative Attachment in Hydrogen

M. Allan and S. F. Wong
Department of Engineering and Applied Science, Mason Laboratory, Yale University, New Haven, Connecticut 06520
(Received 21 September 1978)

We report an electron-beam study of dissociative attachment from excited hydrogen in the energy range of 1–5 eV. A marked dependence of H^-/H_2 and D^-/D_2 cross sections on initial vibrational ($v=0$–5) and rotational ($j=0$–7) states is observed. These results provide a new mechanism to interpret the anomalous H^- density recently observed in a hydrogen plasma.

In dissociative attachment by electron impact, earlier experiments on selected molecules (e.g., O_2, N_2O, CO_2, ...)[1,2] have shown a large increase in cross section with temperature, attributed to the dominant role of vibrationally and rotationally excited molecules. The vibrational effect was first predicted *a priori*[3] and a detailed theoretical study[4] on O^-/O_2 yielded good agreement with experiments.[5] A large rotational effect has also been theoretically predicted[6] on

