Velocity selection of atomic beams in a laser field

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We consider the selection of atoms in the autoionization state under the influence of laser radiation. It is shown in the case of a simple atomic system that the degree of monochromatization, defined by the ratio of the width of the initial to the final velocity distribution, can reach values on the order of 10^2.

1. INTRODUCTION

Obtaining monochromatic atomic beams is of great significance for precision experiments in laser physics and quantum electrodynamics. Studies carried out in recent years on the influence of an intense external electromagnetic field on the autoionization states of atoms open up new possibilities for the velocity selection of atomic beams.[1] The theory of autoionization resonances has been given by Fano.[2] Additional studies of the autoionization states of atoms in a strong electromagnetic field have been carried out by many workers (for example, see Refs. 3–7).

The presence of a discrete level in the continuum leads to interference between the states of the continuum with the discrete state. As a result of such interference, the shape of the autoionization resonance is altered, and new resonances arise, similar to the autoionization resonance. In Ref. 4 it was shown that under certain conditions depending on the strength and frequency of the external field an abrupt change in the photoelectron spectrum is observed near the Fano minimum. This phenomenon is connected with destructive interference of different ionization channels. When the narrowing condition is satisfied, one of the "dressed" states does not decay. This phenomenon is known as "population trapping" and is analogous to the phenomenon of coherent population trapping in a three-level atomic system in a bichromatic laser field and was used in Ref. 8 for laser cooling of atoms. Coherent population trapping in transitions through autoionization states has generated great interest also from the point of view of creating lasers with an uninverted population.[9]

Reference 6 presents a general study of the influence of an external intense electromagnetic field on the autoionization states of atoms. For simple systems (see Fig. 1) the condition for narrowing of the photoelectron spectrum has the following form:

$$\varepsilon=\varepsilon_0=\frac{q}{2}(\Gamma_a-\Gamma_i),$$

where Γ_a and Γ_i are respectively the autoionization width and the ionization width, q is the Fano parameter,[2] and ε is the resonance detuning. The resonance narrowing condition depends on the frequency of the external electromagnetic field and, consequently, on the velocity of the atom, due to the Doppler effect. Depending on the velocity, the narrowing condition is fulfilled for some atoms. Such atoms do not ionize in the laser field, while the remaining atoms will strongly ionize. In the present paper, by considering atomic transitions in the presence of autoionization states, we investigate the possibility of velocity selection of atoms in the presence of a laser field.

2. EFFECTS OF NARROWING OF AUTOIONIZATION RESONANCES

We consider an atom moving with velocity $\mathbf{v}$ in an intense electromagnetic field. For simplicity we assume that $\mathbf{kv}=-kv$, where $\mathbf{k}$ is the wave vector of the electromagnetic field, i.e., the atomic beam and the photon beam are collinear. For atoms moving in response to electromagnetic radiation, it is necessary to replace the frequency of the field ω by $\omega-kv$.

In what follows we consider the simple system shown in the figure. Under the action of an external laser field, transitions from the discrete state to the autoionization state and to the first continuum (the elastic channel) take place. The frequency of the laser radiation is chosen so that the transition from the autoionization state into the second continuum (the inelastic channel) is energetically forbidden. Ionization from the autoionization state to the first continuum always takes place via a two-electron dipole transition, since the autoionization states belong to a mixed term (one in which two or more electrons are simultaneously excited). The oscillator strengths of two-electron transitions are usually of order $10^{-4}-10^{-3}$ (Ref. 10). This transition can be neglected in comparison with the single-electron transition from the discrete state to the first continuum. The degeneracy of the continuum in the orbital quantum number l is removed if the lower discrete state is an s-state.

As the basis wave functions for the discrete state of the atom in a field we choose the quasi-energy wave functions in the resonance approximation. For adiabatic switching-on of the periodic perturbation

$$V(t)=V^+e^{i\omega t}+V^-e^{-i\omega t} \tag{1}$$

these functions have the following form:

$$\Phi_1(t)=\exp\left(-\frac{i}{\hbar}\lambda_1 t\right)(\alpha_1\psi_1+\beta_1\psi_2e^{-i\omega t}),$$

$$\Phi_2(t)=\exp\left(-\frac{i}{\hbar}(\lambda_2-\hbar\omega)t\right)(\alpha_2\psi_1+\beta_2\psi_2e^{-i\omega t}), \tag{2}$$

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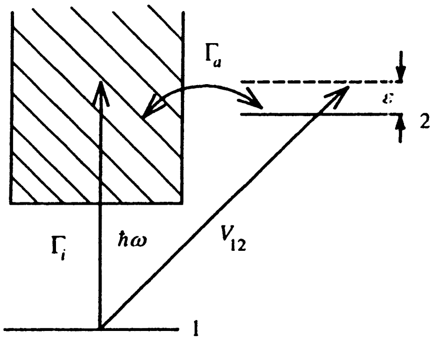

FIG. 1.

where $\lambda^{1,2}$ are the quasi-energies of the atom in the electromagnetic field, and ψ_1 and ψ_2 are the unperturbed wave functions of the free atom,

$$\lambda_1=E_1+\Delta_1,\quad \lambda_2=E_2+\Delta_2+\hbar\omega,$$

$$V_{1,2}=\langle\psi_1|V^+|\psi_2\rangle,$$

$$\Delta_{1,2}=\frac{1}{2}(\varepsilon+\sqrt{\varepsilon^2+4|V_{12}|^2}),\quad \varepsilon=E_2-E_1-\hbar\omega, \tag{3}$$

$$\alpha_{1,2}=\sqrt{\frac{\Delta_{2,1}}{\Delta_{2,1}-\Delta_{1,2}}},\quad \beta_{1,2}=\pm\frac{V_{12}}{|V_{12}|}\sqrt{\frac{\Delta_{1,2}}{\Delta_{1,2}-\Delta_{2,1}}}.$$

When the interaction is switched off, i.e., when $V(t)\to 0$, the quasi-energies λ_1 and λ_2 go over to the energy levels E_1 and E_2 of the free atom, and the functions $\Phi_1(t)$ and $\Phi_2(t)$ go over to the wave functions of the free atom:

$$\Phi_{1,2}\to\psi_{1,2}\exp\left(-\frac{i}{\hbar}E_{1,2}t\right).$$

The energy levels of the states ψ_1 and ψ_2 respectively lie below and above the first ionization threshold.

The Schrödinger equation for this problem has the form

$$i\hbar\frac{\partial\Psi(t)}{\partial t}=[H_0+V(t)+U]\Psi(t), \tag{4}$$

where H_0 is the Hamiltonian of the free atom and U is the Fano "configurational" interaction. We represent the complete solution of Eq. (4) in the form of an expansion

$$\Psi(t)=a_1(t)\Phi_1(t)+a_2(t)\Phi_2(t)+\int dE\psi_E(t)b_E(t), \tag{5}$$

where $\psi_E(t)=\exp(-iEt/\hbar)\psi_E(bfr)$ is the unperturbed wave function of the continuous spectrum of the atom with energy E. Substituting expansion (5) in the Schrödinger equation (4), we obtain a system of differential equations for the coefficients $a_{1,2}(t)$ and $b_E(t)$, which after the Fourier transform

$$a_1(t)=\int d\lambda\exp\left[-\frac{i}{\hbar}(\lambda-\lambda_1-\hbar\omega)t\right]a_1(\lambda),$$

$$a_2(t)=\int d\lambda\exp\left[-\frac{i}{\hbar}(\lambda-\lambda_2)t\right]a_2(\lambda), \tag{6}$$

$$b_E(t)=\int d\lambda\exp\left[-\frac{i}{\hbar}(\lambda-E)t\right]b_E(\lambda),$$

reduce to the following system of algebraic equations for the Fourier coefficients $a_{1,2}(t)$ and $b_E(t)$:

$$(\lambda-\lambda_1\hbar\omega)a_1(\lambda)=\int dEb_E(\lambda)W_{1E},$$

$$(\lambda-\lambda_2)a_2(\lambda)=\int dEb_E(\lambda)W_{2E}, \tag{7}$$

$$(\lambda-E)b_E(\lambda)=W_{1E}^*a_1(\lambda)+W_{2E}^*a_2(\lambda),$$

where

$$W_{1,2E}=\alpha_{1,2}V_E+\beta_{1,2}U_E,\quad V_E=\langle\psi_1|V^+|\psi_E\rangle,$$

$$U_E=\langle\psi_2|U|\psi_E\rangle. \tag{8}$$

This system of equations was studied in the general case in Ref. 6. Here we only give the final expressions for the probability amplitudes for finding the system in the states ψ_1 and ψ_2:

$$C_1(t)=\frac{\exp\left(-\frac{i}{\hbar}\varepsilon t\right)}{x_1-x_2}\left[\left(x_1+\frac{i\Gamma a}{2}\right)\exp\left(-\frac{i}{\hbar}x_1t\right)-\left(x_2+\frac{i\Gamma a}{2}\right)\exp\left(-\frac{i}{\hbar}x_2t\right)\right],$$

$$C_2(t)=\frac{V_{12}\left(1-\frac{i}{2q}\right)}{x_1-x_2}\left[\exp\left(-\frac{i}{\hbar}x_1t\right)-\exp\left(-\frac{i}{\hbar}x_2t\right)\right], \tag{9}$$

where

$$x_{1,2}=-\frac{1}{2}\left[\varepsilon+\frac{i}{2}(\Gamma_a+\Gamma_i)\right]\pm\frac{1}{2}\sqrt{\left[\varepsilon+\frac{i}{2}(\Gamma_a-\Gamma_i)\right]^2+4|V_{12}|^2\left(1-\frac{i}{2q}\right)^2},$$

$$\Gamma_i=2\pi|V_E|^2,\quad \Gamma_a=2\pi|U_E|^2,\quad q=\frac{|V_{12}|}{\sqrt{\Gamma_a\Gamma_i}}. \tag{10}$$

The probabilities of finding the system in the discrete state and the autoionization state are $|C_1(t)|^2$ and $|C_2(t)|^2$, respectively.

It follows from Eqs. (9) and (10) that $C_{1,2}\to 0$ as $T\to\infty$, with the exception of the case

$$\varepsilon=\varepsilon_0=\frac{q}{2}(\Gamma_a-\Gamma_i). \tag{11}$$

When condition (11) holds, one of the roots x_1 and x_2 becomes real, and then the probabilities $|C_1(t)|^2$ and $|C_2(t)|^2$ do not tend to zero as $t\to\infty$. Consequently, when condition (11) is fulfilled the total probability of ionization satisfies $P(T)<1$ in the limit $t\to\infty$. This phenomenon is known as population trapping. Thus, if the frequency and intensity of the external electromagnetic radiation satisfy condition (11), then a discrete state is formed in the continuous spectrum and the resonance narrows.

3. MONOCHROMATIZATION OF ATOMIC BEAMS

When electromagnetic radiation interacts with an atom moving with velocity **v**, the resonance detuning ε will depend on the velocity of the atom due to the Doppler shift of the frequency of the field:

$$\varepsilon = E_2 - E_1 - \hbar\omega\left(1 - \frac{v}{c}\right). \quad (12)$$

Hence it follows that the condition for narrowing of the resonance [Eq. (11)] for given frequency and intensity of the external electromagnetic radiation will be satisfied only for atoms with a certain velocity.

Near the point $\varepsilon - \varepsilon_0$, when the interaction time satisfies the condition $t \gg \hbar/(\Gamma_a + \Gamma_i)$, the expressions for the "survival" probability of the atom will have the form

$$W_{1,2}(t) \approx W_{1,2}(\varepsilon_0)\exp\left[-\frac{\beta}{\hbar}(\varepsilon - \varepsilon_0)^2 t\right], \quad (13)$$

where

$$W_1(\varepsilon_0) \approx \frac{\mathrm{Re}^2 x_1 + \Gamma_a^2/4}{\varepsilon_0^2 + 4|V_{12}|^2 + (\Gamma_a + \Gamma_i)^2/4}$$
$$W_2\left(\varepsilon_0 \approx \frac{|V_{12}|^2(1 + 1/4q^2)}{\varepsilon_0^2 + 4|V_{12}|^2 + (\Gamma_a + \Gamma_i)^2/4}\right. \quad (14)$$

and

$$\beta = \frac{\Gamma_a \Gamma_i}{(q^2 + 1/4)(\Gamma_a + \Gamma_i)^3},$$
$$\varepsilon_0 = E_2 - E_1 - \hbar\omega\left(1 - \frac{v_0}{c}\right). \quad (15)$$

It is clear from expressions (13) and (14) that the main dependence of the probabilities on the atom's velocity is exponential.

After substituting expression (12) in (13), we see that the expressions for the survival probabilities, as functions of thevelocity of the atom take the form

$$W_{1,2}(v) = W_{1,2}(v_0)\exp\left[-\beta\frac{\hbar\omega^2 l}{vc^2}(v - v_0)^2\right], \quad (16)$$

where $W_{1,2}(v_0)$ is the survival probability of an atom moving with velocity v_0 and $l = vt$ is the interaction length of the atom with the radiation. The velocity v_0 is found from expression (12) at $v = v_0$ and the resonance narrowing condition (11):

$$v_0 = c\left\{1 - \frac{1}{\hbar\omega}\left[E_2 - E_1 - \frac{q}{2}(\Gamma_a - \Gamma_i)\right]\right\}. \quad (17)$$

It follows from expressions (12) and (14) that the survival probability of the atom has a maximum at $v = v_0$ and falls off exponentially as the velocity of the atom differs from the value v_0. Such a dependence of the probability on the velocity of the atom makes it effectively possible to obtain monochromatic atomic beams. The width of the velocity distribution (full width at half-maximum of the probability) is given by

$$\Delta v = 2c\sqrt{\frac{v_0 \ln 2}{l\beta\hbar\omega^2} + \left(\frac{c\ln 2}{2l\beta\hbar\omega^2}\right)^2}. \quad (18)$$

4. DISCUSSION OF RESULTS

In what follows we assume that the width of the velocity distribution of the initial atomic beam satisfies $\Delta v \sim v_0$. Then the atomic beam ($\Delta v \ll v_0$) becomes monochromatic under the condition

$$\frac{c^2 \ln 2}{2l\beta\hbar\omega^2} \ll v_0. \quad (19)$$

Hence, taking expression (18) for the width of the velocity distribution of the atomic beam into account, we obtain

$$\Delta v \approx 2c\sqrt{\frac{v_0 \ln 2}{\beta l\hbar\omega^2}}. \quad (20)$$

In very strong and very weak fields condition (19) is violated, since in these cases the parameter β becomes very small. Physically, this means that in very strong fields, when we have $\Gamma_i \gg \Gamma_a$, processes in which the system decays directly prevail over the other processes between the discrete levels, which leads to a breakdown of interference. In weak fields, when $\Gamma_i \ll \Gamma_a$ holds, the ionization time $\tau_i = \hbar/\Gamma_i$ becomes larger than the transit time $\tau = l/v_0$ of the atom across the laser beam, and all the atoms leave the interaction region without ionization.

Let us find the critical value of the laser field for which velocity selection of the atomic beam is possible, i.e., for which condition (19) is fulfilled.

In the case of weak fields, when $\Gamma_i \ll \Gamma_a$ holds, for the parameter β we have (see expression (15))

$$\beta \approx \frac{\Gamma_i}{(q^2 + 1/4)\Gamma_a^2}. \quad (21)$$

Substituting this value of β in condition (19), we obtain

$$\Gamma_i \gg \frac{\Gamma_a^2 c^2 (q^2 + 1/4)\ln 2}{2\hbar\omega^2 v_0 l}. \quad (22)$$

For $v_0 \sim 10^5$ cm/s, $l \sim 10^2$ cm, $\Gamma_a/\hbar\omega \sim 10^{-4}$, and $q \sim 1$, taking into account the condition (22) and the estimate $\Gamma_i \sim \mathrm{Ry}(\mathscr{E}_L/\mathscr{E}_{\mathrm{at}})^2$, where $\mathscr{E}_L$ and $\mathscr{E}_{\mathrm{at}}$ are the field strengths of the laser field and the atomic field, respectively, we obtain the following value for the critical field:

$$\mathscr{E}_{\mathrm{cr}} \sim 10^{-6}\mathscr{E}_{\mathrm{at}} \approx 5 \cdot 10^3 \,\mathrm{V/cm}.$$

In order to obtain near-100% probability of ionization of the atoms not satisfying the resonance conditions, one must use strong laser fields or increase the interaction time. The second approach is preferable to the first since the parameter β decreases in stronger fields and transitions to the continuum (subthreshold ionization[7]), which can join continuum states with energies close to the energy of the autoionization state, become important and destroy interference. This takes place in the presence of the following shift of the ionization threshold:

$$\frac{e^2\mathscr{E}_L^2}{4m\omega^2}\sim E_2-E_i,$$

where E_1 is the energy of the first ionization threshold. Since we have $E_2-E_1\sim\hbar\omega$, it is thus possible to obtain an upper bound on the laser field strength:

$$\mathscr{E}_L\leqslant\left(\frac{\omega}{\omega_{\rm at}}\right)^{3/2}\mathscr{E}_{\rm at}\sim 10^{-2}\mathscr{E}_{\rm at}.$$

As was noted in Ref. 7, the relation $\mathscr{E}_{\rm cr}\sim(\omega/\omega_{\rm at})^{3/2}$ is valid for a short-range interaction of the electron with the remaining ion. If this interaction has a Coulombic nature, the value of the critical field is determined not by the dynamic Stark effect, but by the quasiclassical nature of the transitions near the threshold, which gives a smaller value of the critical field $\mathscr{E}_{\rm cr}\sim(\omega/\omega_{\rm at})^{5/3}$. However, it should be noted that the numerical values of the estimates in both cases are of the same order of magnitude.

Thus, for velocity selection in a laser field it is necessary that the condition

$$10^{-2}\mathscr{E}_{\rm at}\geqslant\mathscr{E}_L\geqslant 10^{-6}\mathscr{E}_{\rm at}.$$

be fulfilled.

Depending on the intensity of the laser field, the quantity β takes its maximum at $\Gamma_i=\Gamma_a/2$. For the same numerical values of the parameters we obtain $\Delta v\sim 10^3$ cm/s for the width of the velocity distribution of the atomic beam, and for its monochromatization, $\Delta v/v_0\sim 10^{-2}$, i.e., the distribution narrows by a factor of 100.

The degree of monochromatization of atomic beams grows as the interaction length l increases. However, for our considerations this length is limited by noninterfering processes: $l<\tau v_0$, where τ is a time determined by noninterfering processes such as the transition from the autoionization state to the second continuum, spontaneous transitions, etc.

If the narrowing condition (11) is fulfilled, then the minimal width is determined by radiative processes. These processes limit the interaction time of an atomic beam in a laser field.

Regarding highly excited atomic states, their lifetimes increase with the principal quantum number n as n^3 (Ref. 11). For example, for $n\sim 30$ we obtain $\tau_{\rm sp}\sim 10^{-4}$ s. Hence it follows that in the case of beams of highly excited atoms this method can be used for efficient velocity selection of atoms with $v_0\sim 10^5$ cm/s.

In conclusion, we note that in the presence of an external electromagnetic field autoionization-like resonances are formed in the continuous spectrum of one-electron atoms.[12,13] The present method of monochromatization can also be applied to such atomic beams.

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