the inverse of the resonance width, is given by

$$T \sim \sqrt{\sigma}/\bar{v} \sim n^{*2}/\bar{v}^{3/2}. \quad (4)$$

Clearly the observed values of both σ and T exhibit the same n^* dependences as Eqs. (3) and (4). Our calculations indicate values of $\sigma = 3\times 10^8$ Å^2 and $T = 10^{-9}$ s for the 18s state in good, somewhat fortuitous, agreement with the observed values shown in Fig. 3. It is interesting to note that according to our calculations the (0, 1) and (1,0) resonances should not occur for a spinless system but occur here because of the spin-orbit coupling.

In conclusion we have observed that the long-range dipole-dipole interaction leads to enormous cross sections and the long interaction times requisite for sharp resonances in the collision cross section. The long duration (1 ns) of the collisions may open the way to such interesting investigations as the introduction of perturbations in mid collision.

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Confluence of Bound-Free Coherences in Laser-Induced Autoionization

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We solve exactly the problem of double configuration mixing (Coulombic and radiative) in a "Fano model" atom, and predict a new confluence of coherences in laser-induced autoionized electron spectra.

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Close-coupling or dressed-state methods are used in theoretical physics when two states are very strongly mixed by a perturbing interaction. In essence, such methods amount to *ad hoc* but exact rediagonalizations of a particularly sensitive part of the Hamiltonian after the eigenstates are first determined in a "natural" but eventually inappropriate basis. This rediagonalization is trivial only if the states are discrete. Fano's solution to the discrete-plus-continuum rediagonalization is central to his classic discussion of autoionization.[1]

In this note we consider the situation in which the "natural" basis is doubly inappropriate because two physically distinct strong mixings are present, and both of them mix the same group of discrete and continuum states. This situation presents a new problem, which we have solved exactly. Our doubly rediagonalized solutions may be of interest in their own right, but they also indicate the existence of an unexpected confluence of bound-free coherences that may be observable in laser-induced excitation of autoionizing resonances.[2]

The situation we have in mind is indicated by the "Fano model" atomic-energy-level diagram in Fig. 1. The figure shows that a bound state in one electronic configuration, labeled $|1\rangle$, can overlap states in the continuum of another configuration, labeled $|\omega\rangle$. The Coulombic interaction between the configurations mixes these states strongly, leading to the creation of an autoionizing resonance.[1] At the same time, if the radiative excitation from a lower bound state, labeled $|0\rangle$, is strong enough, then state $|0\rangle$ is thoroughly mixed into the resonance by photon exchange.

The Hamiltonian for the system described can be written (with $\hbar = 1$)

$$\hat{H} = \hat{H}_0 + \hat{H}_1 + \hat{H}_\omega + \hat{V}_C + \hat{V}_r \tag{1}$$

where the various parts of $\hat{H}$ are the bare energies

$$\hat{H}_0 = E_0|0\rangle\langle 0|, \quad \hat{H}_1 = E_1|1\rangle\langle 1|, \quad \hat{H}_\omega = \int d\omega\, \omega|\omega\rangle\langle\omega|, \tag{2}$$

and the interaction energies: Coulombic,

$$\hat{V}_C = \int d\omega\, V_1(\omega)|1\rangle\langle\omega| + \text{H.c.}; \tag{3a}$$

and radiative,

$$\hat{V}_r = V_{01}\exp(i\omega_L t)|0\rangle\langle 1| + \int d\omega\, V_0(\omega)\exp(i\omega_L t)|0\rangle\langle\omega| + \text{H.c.} \tag{3b}$$

Here ω_L is the (laser) photon frequency responsible for the radiative excitation.

The first rediagonalization was accomplished by Fano.[1] The new states, which we denote by round-bracket vectors, $|\omega)$, are eigenstates of $\hat{H}_{1\omega C} = \hat{H}_1 + \hat{H}_\omega + \hat{V}_C$. As Fano showed, the matrix elements of a transition operator (called T by Fano, and given by $\hat{V}_r$ in the present case) between an initial state $|0\rangle$ and continuum states in the old and new bases are related by

$$\langle 0|\hat{V}_r|\omega) = \langle 0|\hat{V}_r|\omega\rangle e^{i\varphi}\frac{\epsilon(\omega)+q}{\epsilon(\omega)-i}. \tag{4}$$

The absolute square of Eq. (4) is exactly Fano's Eq. (21); φ is an arbitrary phase, $\epsilon(\omega)$ is Fano's reduced energy,

$$\epsilon(\omega) \equiv [\omega - E_1 - F(\omega)]/\gamma_0, \tag{5}$$

and q is the well-known asymmetry parameter. We will hereafter ignore the small shift $F(\omega)$.

We express $\hat{V}_r$, using the new round-bracket basis:

$$\hat{V}_r = \int d\omega\, \Omega_\omega \exp(i\omega_L t)|0\rangle(\omega| + \text{H.c.} \tag{6}$$

A comparison of the matrix elements of (6) with expression (4) shows that $\Omega_\omega \exp(i\omega_L t) = \langle 0|\hat{V}_r|\omega)$ or

$$\Omega_\omega \exp(i\omega_L t) = (q+i)e^{i\varphi}\langle 0|\hat{V}_r|\omega\rangle\left[\frac{1}{\epsilon(\omega)-i} + \frac{1}{q+i}\right]. \tag{7}$$

For convenience we denote by $\Omega_\omega \exp(i\omega_L t)/(4\pi\gamma_0)^{1/2}$ the collection of parameters multiplying the square bracket in (7), adjusting φ so that Ω_0 is real. We also introduce a large ($\sigma \gg 1$) width into the flat background term:

$$\frac{1}{q+i} \to \frac{1}{q+i}\frac{i\sigma}{\epsilon(\omega)+i\sigma}.$$

The second rediagonalization is less simple than the first. The sinusoidal oscillation of the laser field must be accounted for, and Ω_ω need not be a smooth function of ω if γ_0 is small. Nevertheless, we have obtained an exact solution. If the time-dependent Schrödinger wave vector is expanded in the round-bracket basis: $|\psi(t)\rangle = \alpha(t)|0\rangle + \int d\omega\, \beta_\omega(t)|\omega)$, then the equations for α and β_ω (in the rotating frame, assuming a step turn-on of the laser and taking $E_0 = 0$ for convenience) are

$$\dot{\alpha} = -i\int d\omega\, \Omega_\omega \beta_\omega, \tag{8a}$$

$$\dot{\beta}_\omega = -i(\omega - \omega_L)\beta_\omega - i\Omega_\omega{}^*\alpha. \tag{8b}$$

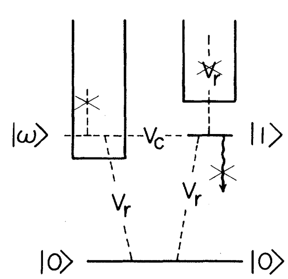

FIG. 1. Simplified atomic level scheme, showing the Coulombic and radiative interactions, V_C and V_r. The crosses show that spontaneous radiative decay and free-free transitions are not considered in the model.

The Laplace transforms of $\alpha(t)$ and $\beta_\omega(t)$ are easily found. For example,

$$\frac{1}{\tilde{\alpha}(z)} = z + \int d\omega \frac{|\Omega_\omega|^2}{z + i(\omega - \omega_L)}, \qquad (9)$$

which may be familiar as the resolvent $\mathcal{H}(z)$ for Schrödinger's equation; i.e., $\tilde{\alpha}(z) = 1/\mathcal{H}(z)$. This result is generally important because it is well defined for any integrable coupling function Ω_ω. In the present case, where the first rediagonalization has led to expression (7) for Ω_ω, we obtain the following explicit formula for the resolvent (in the limit that the excitation is many many times γ_0 above the ionization edge):

$$\mathcal{H}(z) = z + \tfrac{1}{4}\Omega_0^2[(z + i\Delta + \gamma_0)^{-1} + (\bar{\gamma}_0/\gamma_0)(1+q^2)^{-1}(z + i\Delta + \bar{\gamma}_0)^{-1} - 2\bar{\gamma}_0(1+iq)^{-1}(z + i\Delta + \gamma_0)^{-1}(z + i\Delta + \bar{\gamma}_0)^{-1}], \quad (10)$$

where $\Delta = E_1 - \omega_L$ and $\bar{\gamma}_0 = \sigma\gamma_0$.

Although $\mathcal{H}(z)$ is a rational function of only third degree, for practical purposes the time dependences are best obtained by numerical Laplace inversion. An example is shown below. However, one of the most interesting features of our result can be obtained completely analytically. The spectrum $W(\omega)$ of outgoing free electrons with energy ω is given exactly by $|\beta_\omega(t)|^2$. (Note that this is the distribution of outgoing electron energies for *fixed* laser frequency ω_L. It is not a generalization of the well-known Fano profile.) In the long-time limit, when essentially all of the electrons have been ionized, the simple relation

$$W(\omega) \to \left|\frac{\Omega_\omega}{\mathcal{H}(-i(\omega - \omega_L))}\right|^2$$

gives a complicated but completely explicit formula for the spectrum, that we will present elsewhere.[3]

Here we will restrict ourselves only to the most interesting features of the spectrum. These occur because the two rediagonalizations can interfere with each other and because the degree of interference can be changed by changing the laser intensity. In Fig. 2 these interferences are exhibited. The small inset in the corner shows the long-time photoelectron spectrum for a very large q value and three values of laser power. The splitting is a pure Autler-Townes effect.[4] It shows that the Coulombic configuration mixing can be dominated by the strong radiative coupling. The three spectral curves can be interpreted in the time domain. The single-peaked curve accompanies smooth monotonic leakage of population from $|0\rangle$ into the continuum (as expected for low Ω_0); whereas the double-peaked curves indicate the presence of strong population oscillations between $|0\rangle$ and the distorted continuum (as appropriate for $\Omega_0 > \gamma_0$). The splitting of photoelectron spectra has been discussed in other contexts recently[5] but has not yet been observed (see also Fig. 3 below).

The main part of Fig. 2, however, shows new behavior. In this case we have $q = 1$, indicating a strong Fano asymmetry with its maximum at -1 on the normalized energy axis. The dashed vertical line indicates that the laser is tuned to this maximum. The first notable feature is the inelasticity of the spectrum. The spectral peaks (labeled by values of Ω_0^2 in units of γ_0^2) are not sharp, even though the laser is monochromatic, and none of the peaks are located at the laser frequency. The inelasticity increases with laser power. Secondly, the splitting of the Autler-Townes peaks (the figure shows only the right-hand peaks) is greatly different from that shown in the inset curves. As the laser power is *in-*

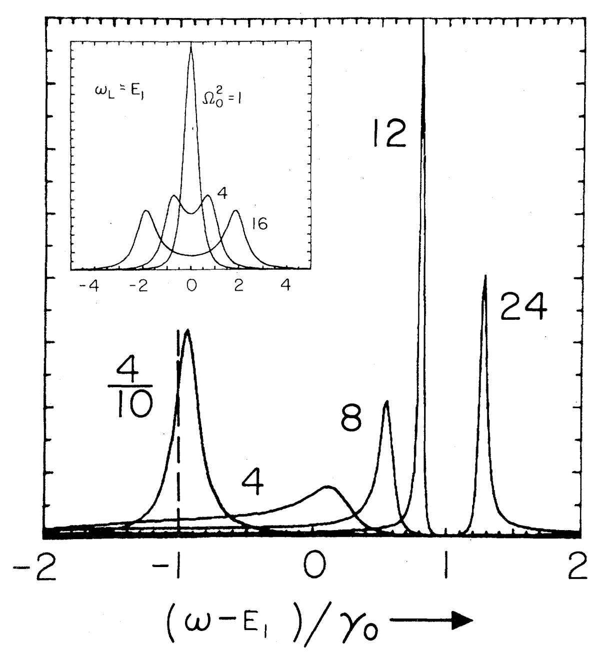

FIG. 2. Long-time electron spectra for several laser intensities, labeled by the value of Ω_0^2 (in units of γ_0^2). The inset shows spectra for $q >> 100$ and the main figure shows spectra for $q = 1$. The dashed vertical line at -1 shows the (resonant) laser frequency. The position of the confluence is evident at +1 on the normalized energy axis.

creased (curves labeled 4, 8, 12) a remarkable line *narrowing* occurs. This narrowing reverses for still higher powers, and the shape of the peaks changes, with a long tail to the right of the last peak, but a tail to the left of the other peaks. There is zero spectral amplitude at $\omega = E_1 + \gamma_0$.

These new features in the photoelectron spectrum are apparently explained by saying that a confluence of coherences occurs at $\omega = E_1 + \gamma_0$. This is exactly the position of the Fano minimum associated with the Coulombic rediagonalization. One can argue heuristically that the autoionization mechanism provides a channel by which electrons originally in level $|0\rangle$ can reach the continuum, and thus autoionization acts as a probe of the Autler-Townes effect. However, with increasing laser power, as the Autler-Townes peak moves into the Fano minimum the effectiveness of the radiative matrix element decreases, reducing the throughput of the autoionization channel. As the channel closes, the lifetime of the bound state increases dramatically, explaining the sharp narrowing in linewidth.

The critical value of laser intensity, corresponding to exact confluence of the right-hand Autler-Townes peak with the Fano minimum, can be estimated as follows. The Autler-Townes splitting is normally (see the high-q case in the inset) equal to the Rabi frequency Ω_0. In Fig. 2 the "half-splitting" corresponding to the separation between the laser tuning (at -1) and the Fano minimum (at $+1$) suggests that $\frac{1}{2}\Omega_0 = 2$ gives the critical value. This corresponds to $\Omega_0^2 = 16$, which is in very good agreement with the values around the confluence in Fig. 2.

Our solution (10) is exact, and predicts the existence of specific new features in the photoelectron spectrum. Thus it is important to state the limitations of our model. Spontaneous radiative decay has been assumed absent, and the laser has been assumed purely monochromatic. The second assumption can be removed,[3] and the effect is to broaden the spectral peaks, as expected. Additional radiative interactions with higher continuum levels are ignored. Such interactions are not expected to alter our predictions, unless higher autoionizing resonances are assumed present.[6] The results given in Fig. 2 are all based on the long-time limit, and actual experiments would be done in short times. As an indication that our results do not appreciably distort the finite-time picture, we show in Fig. 3 a graph of the time dependence of the population of level $|0\rangle$. The oscillations in the strong-field

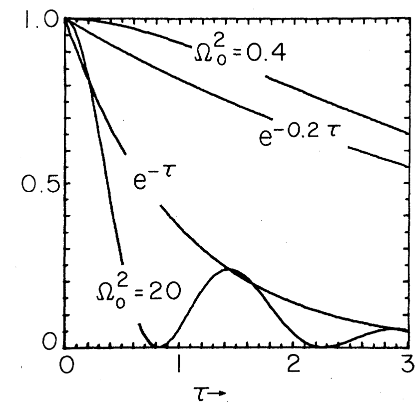

FIG. 3. Exact time dependence of the population of state $|0\rangle$ is shown in the two curves labeled by Ω_0^2 (in units of γ_0^2). The two exponential curves show that for low Ω_0 the population leaks out at a rate approximated by $\Omega_0^2/2\gamma_0$ (which is the one-photon low-power ionization rate), and for high Ω_0 at the rate γ_0 (which is the width of the autoionizing resonance).

curve are further indications of the Autler-Townes splitting of the autoionizing resonance. We should also mention that the "high" laser powers needed to observe bound-free coherence may not be very high at all, as new methods have been reported recently[7] for creating autoionizing resonances as narrow as a few hundred MHz and with $q < 10$.

Finally, it is interesting to note that our heuristic explanation of the confluence of coherences cannot be fully correct. The throughput of the autoionization channel can also be reduced simply by reducing the laser power (weakening the radiative coupling). The automatic result is to increase proportionately the lifetime of the bound state, but one does not thereby narrow the autoionized electron spectrum below the low-field value $\Omega_0^2/2\gamma_0$. The confluence of radiative and Coulombic interactions is apparently more subtle, and its most interesting features are intrinsically strong coupling in nature.

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