Ramsey Interference Fringes in Radiatively Assisted Collisions of K Rydberg Atoms

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When resonant collisional energy transfer occurs in a radiation field of frequency comparable to the inverse of the collision time, Ramsey interference fringes are observed in the cross section. At low frequencies the fringes are phase dependent, but at high frequencies they are not. The fringes may be understood with a quasistatic field description, which ties together the low- and high-frequency regimes.

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Radiatively assisted collisions, those in which two atoms colliding in a radiation field absorb or emit photons during the collision, have primarily been studied in the regime in which the frequency of the field ν is high compared to the inverse of the collision time τ, i.e., $\nu \gg 1/\tau$ [1]. In this case the collisions are well understood as collisions of atoms dressed by a monochromatic field. At the other extreme, $\nu \ll 1/\tau$, the collisions occur in the presence of a quasistatic field, the value of which depends on the phase of the radiation field at which the collision occurs [2,3]. In contrast to these two extreme cases, it is not *a priori* evident how to describe collisions in which $\nu \approx 1/\tau$, nor is it straightforward to study such collisions experimentally, for the phase of the radiation field at which the collision occurs must be controlled.

Resonant energy-transfer collisions between Rydberg atoms [4] present an excellent opportunity to study such collisions. Here we report the experimental study of radiatively assisted resonant collisional energy transfer between K Rydberg atoms in the regime $\nu = 1/\tau$. Specifically, we have used velocity-selected atoms which have inherent collision times of ≈ 1 μs and are only allowed to collide during a ≈ 1-μs interval. Phase locking a radio-frequency (rf) field of frequency ≈ 1 MHz to this interval provides the requisite phase control. The measurements show the evolution from the phase-dependent low-frequency regime to the phase-independent high-frequency regime. The most striking aspect of the results is that, when $\nu \approx 1/\tau$, the cross sections exhibit quantum interference fringes, the number of which depends on the rf field amplitude. We present a description of the collisions which shows that the fringes are similar to those observed in a Ramsey separated-oscillatory-field experiment [5], to the Stuckelberg oscillations [6] observed in differential scattering cross sections, and to the fringes observed using isolated atoms in combined static and rf fields [7]. This description is related in a general way to the evolution with frequency of multiphoton transitions of isolated atoms [8] and it allows us to connect in a transparent way the low- and high-frequency regimes.

The experimental approach is straightforward. As shown in Fig. 1, a thermal beam from an oven intersects a rotating chopper wheel which passes a 10-μs pulse of atoms. 15 cm downstream and 200 μs later the atoms are excited from the ground $4s_{1/2}$ state to the $4p_{1/2}$ state by a 5-ns laser pulse, which ensures that only a narrow velocity distribution of atoms is excited [3]. A focusing hexapole electromagnet increases the number of colliding atoms by a factor of 10 [9]. Not shown in Fig. 1 is a magnetic shield which reduces the magnetic field in the interaction region to less than 5 mG.

Using two additional pulsed dye lasers, we further excite the velocity-selected K $4p_{1/2}$ atoms to the $29s_{1/2}$ and $27d_{3/2}$ Rydberg levels in the presence of a static electric field of ≈ 6.5 V/cm. As shown in Fig. 2, in this field the $29p_{1/2}$-$29s_{1/2}$ and $27d_{3/2}$-$28p_{1/2}$ energy intervals are iden-

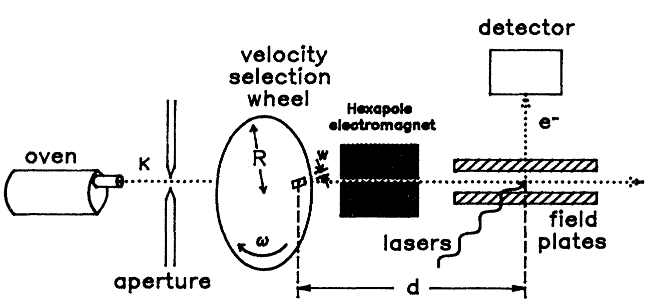

FIG. 1. Experimental apparatus.

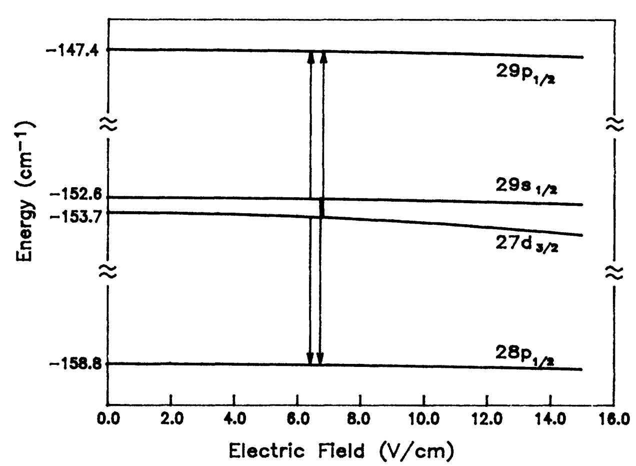

FIG. 2. Energy-level diagram for the Rydberg states of K involved in the resonant energy-transfer collisions being studied. The two transitions shown have been separated for clarity; they are degenerate in electric field.

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tical, and the initially excited atoms can resonantly transfer energy via a dipole-dipole interaction to yield one atom in the $29p_{1/2}$ state and the other in the $28p_{1/2}$ state.

The allowed collision interval τ begins when the Rydberg levels are populated and ends 0.8 μs later with the application of a 50-ns rise-time detuning pulse to the lower field plate. Shortly after the detuning pulse, a high-voltage pulse is applied to the lower plate to selectively ionize the $29p_{1/2}$ atoms and accelerate the resulting electrons into a microchannel-plate detector. This signal is recorded as the static electric field E_s is swept through the field of the collisional resonance over many laser pulses. A radio-frequency voltage is applied continuously to the upper field plate and the laser pulses are synchronized to the rf phase, thus phase locking the rf field to the allowed collision interval.

Collisional resonances observed in the presence of a phase-locked 0.75-MHz, 0.21-V/cm rf field ($\nu=0.6/\tau$) are shown in Fig. 3. The rf field is given by $E_{\rm rf}\cos(\omega t+\phi)$, where $t=0$ is the center of the allowed collision interval, 0.4 μs after the laser pulses. In Fig. 3(a) we show the 1.4-MHz-wide resonance in the absence of an rf field, while the resonances of Figs. 3(b) and 3(c) have phases of $\phi=0$ and $\pi/2$, respectively. The data agree with the dashed line shapes obtained by numerically integrating the Schrödinger equation. In particular, for $\phi=0$, in Fig. 3(b), one can see that the largest peak of the resonance is shifted to a lower electric field by an amount equal to the rf amplitude, and subsidiary peaks are observable at higher static electric fields. We note that the spacing between the peaks is more than twice the 0.023-V/cm spacing expected for a 0.75-MHz-photon-assisted resonance. In contrast to the $\phi=0$ case, for $\phi=\pi/2$, shown in Fig. 3(c), no distinct peaks are observable.

We can describe these collisions in terms of interacting molecular states in a quasistatic field. The two molecular states are the initial state Ψ_i, composed of the $29s_{1/2}$ and $27d_{3/2}$ states, and the final state Ψ_f, composed of the $29p_{1/2}$ and $28p_{1/2}$ states. To a good approximation the energy of the final state W_f is field independent, but the initial-state energy W_i has a linear Stark shift. Near the resonance field E_R at which $W_i=W_f$, the energy difference between the initial and final states is given by the linear approximation,

$$W_i-W_f=k_{\rm eff}(E-E_R)\,, \qquad (1)$$

where $k_{\rm eff}=dW_i/dE|_{E_R}$ has the value 32.5 MHz/(V/cm). There are two requirements for a collisional transition to occur. First, the energy resonance condition $W_i=W_f$ must be met, and second, the dipole-dipole interaction, integrated over the collision time, must be strong enough to induce the transition. In the absence of an rf field, the resonance condition can be met for the full 0.8-μs interval, leading to the simple 1.4-MHz-wide resonance seen in Fig. 3(a).

Consider now the case in which an rf field of $\phi=0$ and

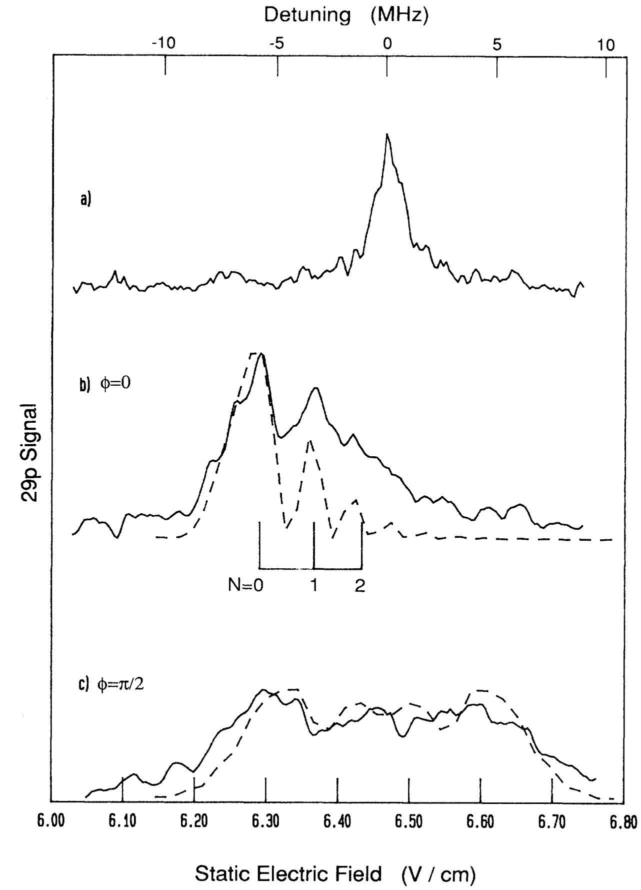

FIG. 3. The experimental (——) and calculated (---) line shapes of the $n=29$ radiatively assisted resonance of K as functions of the static field E_s. (a) The unperturbed resonance with no rf field present, FWHM$=1.4$ MHz. (b),(c) The resonance in the presence of a 0.75-MHz, 0.21-V/cm rf electric field and a phase of (b) $\phi=0$, (c) $\phi=\pi/2$. Indicated below the spectra are the calculated positions of the peaks using the $\Phi=2\pi N$ condition.

$\nu=0.75$ MHz is added. When $E_s+E_{\rm rf}=E_R$, the resonance condition is met, at $t\approx 0$, but not for the entire 0.8-μs allowed collision interval. Thus we expect to see the collisional resonance at $E_s=E_R-E_{\rm rf}$, but, due to the shorter time on resonance, it should be broader than the resonance with no rf field. As shown by Fig. 3(b), there is a broad peak at $E_s=E_R-E_{\rm rf}$; however, there are also smaller peaks at $E_s>E_R-E_{\rm rf}$.

The origin of the smaller peaks becomes apparent by examining Fig. 4(a). When $E_s>E_R-E_{\rm rf}$, the resonance condition is met twice, at $t=\pm t'$, i.e., there are two interaction periods, just as in the Ramsey separated-oscillatory-field method. In the first interaction, at $t\approx -t'$, a coherent superposition of Ψ_i and Ψ_f is formed. Between the interactions at $-t'$ and t', the Stark shift of Ψ_i alters its phase relative to that of Ψ_f. The accumulated phase

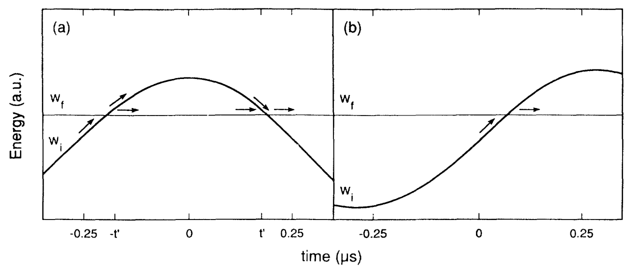

FIG. 4. The initial- and final-state energies, W_i and W_f, in a 0.75-MHz field with phases (a) $\phi=0$ and (b) $\phi=\pi/2$. In both cases, $E_s > E_R - E_{\rm rf}$ and the allowed collision time τ is 0.7 μs. As the static field is scanned, when $\phi=0$ the single interaction period at $t=0$, when $E_s = E_R - E_{\rm rf}$, evolves into two interaction periods at $t=\pm t'$, when $E_s > E_R - E_{\rm rf}$, as shown in (a). The transition amplitudes interfere constructively when the total phase shift of Ψ_i between $-t'$ and t' is $2\pi N$. In (b) only one interaction period occurs, irrespective of E_s.

difference,

$$\Phi = \int (W_i - W_f)dt = \int k_{\rm eff}(E - E_R)dt \,, \tag{2}$$

is simply proportional to the area between the two curves of Fig. 4(a). If $\Phi = 2\pi N$, where N is an integer, the transition continues in the second interaction period as if the two interaction periods were not separated in time, i.e., there is constructive interference. On the other hand, if $\phi = 2\pi(N+\frac{1}{2})$, the transition amplitude of the second interaction period cancels that of the first, i.e., there is destructive interference, and no transition occurs. For a given $E_{\rm rf}$ we expect to see a sequence of peaks corresponding to $N=0,1,2,\ldots$ as we sweep the static field. In Fig. 3 we show the computed locations of the $N=0$, 1, and 2 peaks, which are in good agreement with the data.

If $\phi=\pi/2$, at 0.75 MHz there is only one time at which $W_i = W_f$, as shown by Fig. 4(b). As a result, interference fringes do not occur and only one resonance extending from $E_R - E_{\rm rf}$ to $E_R + E_{\rm rf}$ is observed, as shown in Fig. 3(c). The collisional resonances for phases of $\phi=\pi$ and $-\pi/2$ are the mirror images, in static field, of the $\phi=0$ and $\pi/2$ cases, respectively.

To show the effect of increasing the frequency to $\nu = 1.18/\tau$ we show in Fig. 5(a) the resonances obtained with $E_{\rm rf} = 0.31$ V/cm, $\nu = 1.48$ MHz, and $\phi = 0$. The experimentally observed resonances match the spectrum predicted using the $\Phi = 2\pi N$ requirement and direct numerical integration of the Schrödinger equation. When $\nu = 1.48$ MHz and $\phi = \pm\pi/2$, $W_i = W_f$ at three times, and the amplitudes from the different interaction periods interfere, producing the interference fringes shown in Fig. 5(b). As shown in Fig. 5(b), the observed and computed spectra are in reasonable agreement. At $\nu = 1.48$ MHz the phase is not very important, and when $\nu = 4.0$ MHz we have reached the high-frequency limit and are unable to detect a phase dependence.

It is instructive to apply this model to the high-

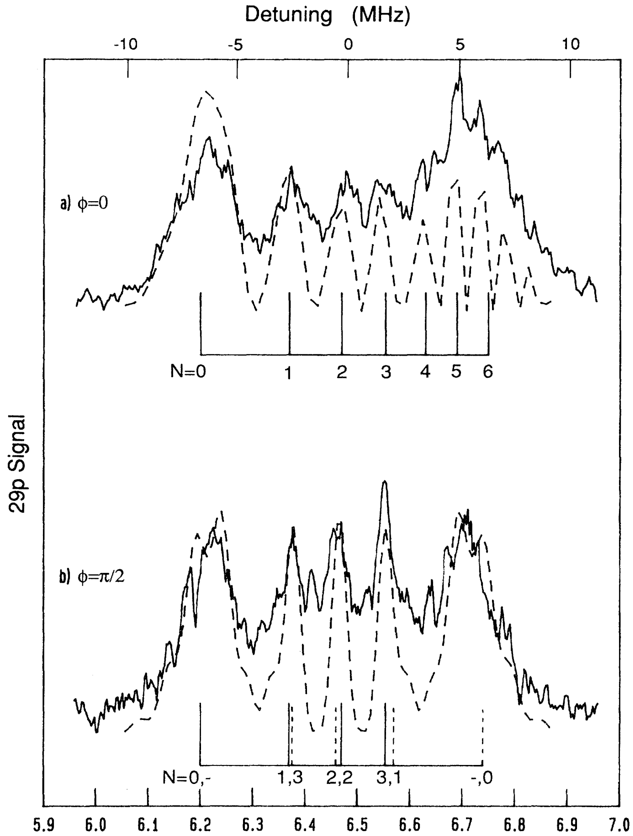

FIG. 5. The experimental (——) and calculated (---) line shapes as functions of the static field E_s in a 0.31-V/cm, 1.48-MHz rf field of phases (a) $\phi=0$ and (b) $\phi=\pi/2$. The allowed collision time is 0.8 μs. The calculated fringe positions using $\Phi=2\pi N$ are indicated below the spectra. In (b) the fringe positions due to interference between the first and second (solid vertical marker) and the second and third (dashed marker) interactions are shown.

frequency case in which there are many crossings of W_i by W_f, the effects of which must add coherently. For constructive interference, after each rf cycle Ψ_i and Ψ_f must be the same, or $\Phi = 2\pi N$ when integrated over one rf cycle. Since the integral of the rf field over a cycle is zero, this requirement implies that the resonances will be found at

$$E_s = N\omega/k_{\rm eff} + E_R \,, \tag{3}$$

i.e., at the static fields corresponding to an integer times the field detuning of one rf photon, the same result as obtained in the high-frequency theory [2]. The strength of the N-photon resonance depends upon the accumulated phase in each of the two parts of the cycle when $W_i > W_f$ and $W_i < W_f$. When these phases are both integral multiples of 2π, the resonances are strong. For small N and large rf fields this requirement leads to an oscillation in the intensity of the Nth resonance as $|\cos(k_{\rm eff}E_{\rm rf}/\omega$

$-\phi_0)|$ which has the same period as does the Bessel-function expression of the high-frequency theory [2].

In conclusion, when the added radiation field is of frequency $\nu \approx 1/\tau$ the radiatively assisted collisions exhibit Ramsey fringes, which can be understood in terms of a quasistatic field description that bridges the gap between the high- and low-frequency regimes.

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