Ionization of Rydberg Atoms by a Circularly Polarized Microwave Field

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We report the first observation of the ionization of Rydberg atoms by a circularly polarized 8.5-GHz microwave field. The observed threshold field for ionization is $E=1/16n^4$, a value higher than that required for ionization by a linearly polarized microwave field. The origin of this threshold can be understood by transforming to a frame rotating at the microwave frequency. In the rotating frame no transitions occur, and level mixings alone are responsible for the ionization.

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Optical multiphoton experiments have established that, to achieve the same ionization rate, higher intensities are required for circularly than linearly polarized light and that the difference in the required intensities increases with the order of the process.[1] The observed difference between the two polarizations is in accord with theory[2] and can be understood in a straightforward way.[3] Consider, for example, multiphoton ionization of an atom initially in an s state. With circularly polarized light the electron's orbital angular momentum, l, must increase by one with the absorption of each photon. On the other hand, with linearly polarized light the absorption of each photon can change l by ± 1 (except when passing through an s state), increasing the number of paths and potential resonances en route to ionization and the efficiency of the process.

Rydberg atoms in microwave fields clearly display high-order multiphoton processes,[4,5] suggesting that the difference between circular and linear polarization might be dramatic in this case. In addition, the experiments have shown that these processes can be thought of not only as multiphoton processes, but also as being driven by the time variation of a quasistatic field,[5] raising the question of the connection of a slowly rotating field to a static field. With these two questions in mind we have begun to study microwave ionization by a circularly polarized field. Here we report the first experimental study and analysis of this problem.

In our experiment Na atoms from a resistively heated oven are collimated into a beam which passes between the mirrors of a Fabry-Perot microwave cavity. At the center of the cavity the atoms are excited from the ground $3s$ state through the $3p$ state to a Rydberg state of $24<n<56$ by two pulsed dye laser beams. Here n is the principal quantum number. Depending upon its strength, the microwave field may or may not ionize the Rydberg atoms. About 1.6 μs after the laser pulses, a 3-kV pulse is applied to a plate below the cavity, which produces a field of 230 V/cm at the location of the Rydberg atoms, accelerating any ions formed through a grid in a plate above the cavity to a microchannel plate detector. The signal from the detector is recorded with a gated integrator.

The Fabry-Perot microwave cavity consists of two 16-cm radius-of-curvature brass mirrors 10.7 cm in diameter separated by 4.5 cm on their axis. We operate the cavity on the TEM_{002} mode at a frequency of 8.5 GHz. To produce the circularly polarized microwave field we feed orthogonally polarized microwave fields 90° out of phase into the cavity through 8.5-cm-diam irises in the two mirrors. The waveguides connected to the two mirrors are perpendicular to provide the horizontally and vertically polarized fields. The nominal 8.5-GHz resonances for the two polarizations are offset by 2 MHz, well within the linewidths of the modes, which have Q's of 2000 and 2100. The differences in the frequencies and Q's of the modes are due to the imperfections in the circular symmetry of the cavity and its surroundings.

The microwave power originates in a Hewlett-Packard (HP) 8350B sweep oscillator with an 83550-A plug in, and passes through a Triangle Microwave 2-GT-41 voltage controlled attenuator before being amplified by a Litton 624 pulsed traveling-wave-tube amplifier. After the amplifier the power is split between two arms feeding the two mirrors. In one of the two arms is an HP X885A phase shifter followed by an HP X382A precision variable attenuator, and in the other a fixed 3-dB attenuator. We measure the power at one input to the cavity with a HP 432A power meter and use the known mode geometry to calculate the field, with an uncertainty of $\pm 12\%$.

To produce initially a circularly polarized field we used two weakly coupled probes to monitor the microwave fields polarized at $\pm 45°$ from the vertical. We adjusted the phase shifter and attenuator to extinguish one of these polarizations. The subsequent introduction of a phase shift of 90° produced a field adequately circularly polarized to see clearly the difference between ionization by circularly and linearly polarized fields. In later experiments we have removed the probes and simply used the ionization signal to adjust the polarization.

We have excited the atoms both before and during the microwave pulse, and in either case we observe the microwave ionization signal as the microwave power is attenuated with the voltage controlled attenuator. In Fig. 1 we show the microwave ionization signals obtained

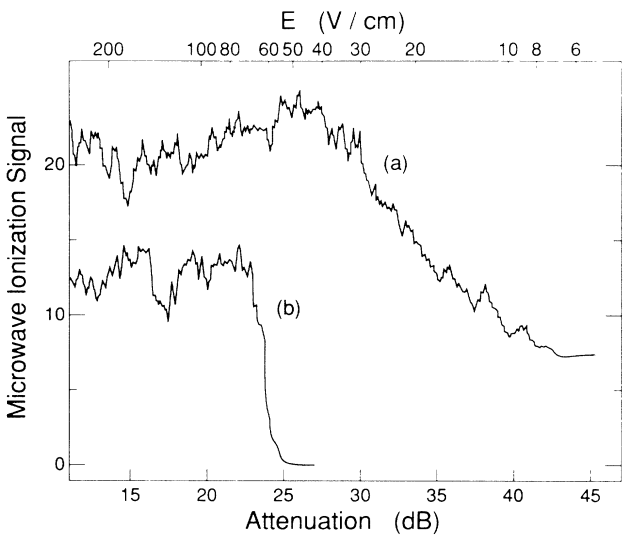

FIG. 1. Microwave ionization signals as a function of microwave attenuation when the Rydberg atoms are excited in the microwave field at the energy of the 46*d* state; curve *a*, linearly polarized microwaves and curve *b*, circularly polarized microwaves. The two traces are offset vertically by eight units for clarity. For reference a microwave field scale is shown as well.

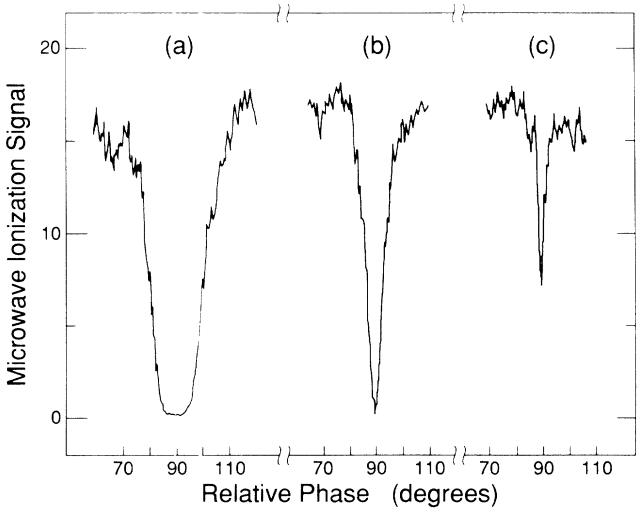

FIG. 2. Microwave ionization signal as a function of relative phase between the horizontal and vertical fields when the atoms are excited to the energy of the 46*d* state and the attenuation is set between the linear- and circular-polarization ionization thresholds. Specifically, scans of the phase are shown for microwave fields of (a) 34.6 V/cm, (b) 53.5 V/cm, and (c) 61.5 V/cm.

when we excite the atoms to the energy of the zero-field 46*d* state in the presence of both linearly and circularly polarized microwave fields. It is apparent that ionization requires a larger field when circular rather than linear polarization is used.

The two traces of Fig. 1 suggest the most sensitive way of adjusting for circular polarization. The horizontal and vertical fields can be adjusted to the same amplitude by requiring that they separately have the same linear-polarization ionization thresholds. Using both fields, with the same amplitudes, we set the attenuation between the linear- and circular-polarization thresholds of Fig. 1 and sweep the phase, which produces a sharp decrease in the ionization signal at the correct phase for circular polarization, as shown in Fig. 2. Not surprisingly, the microwave ionization signal varies more sharply with phase when the attenuation is set nearer to the circular-polarization threshold as shown in Fig. 2. More fundamentally, Fig. 2 shows that a small ellipticity in the polarization dramatically reduces the field required for ionization. Similarly, once circular polarization is obtained, increasing or decreasing the amplitude of either linear-field component increases the ionization signal.

In Fig. 3 we show the n dependence of the ionization threshold fields obtained when the Rydberg atoms are excited in the microwave field for linearly and circularly polarized fields. As shown, for $n < 40$ the field required for linear polarization is $1/3n^5$ (atomic units), as previously observed.[6] For $n > 40$ the thresholds become broader and move to higher fields, as observed previously.[7] In contrast, the field required for circular polarization is $1/16n^4$ (atomic units), approximately the same as for a nonhydrogenic atom in a static field.

When we excite the atoms in zero field and then turn on the microwave field, for linear polarization and for high-n states with circular polarization we obtain the threshold fields shown in Fig. 3. However, for circular polarization the $n < 30$ states ionize at fields between $1/3n^5$ and $1/16n^4$. Since the low-n states are very sensitive to a small ellipticity in the polarization, we attribute the reduction in threshold fields to ellipticity in the polarization as the two cavity modes with different Q's fill. However, the reduction might also be due to dynamic Landau-Zener transitions.[8]

To understand ionization by a circularly polarized field we begin by recalling the difference between linearly and circularly polarized fields. With linear polarization the axis of quantization is along the field, zero-field states of the same m are coupled, and m remains a good quantum number. Here m is the azimuthal orbital angular momentum. With circular polarization the axis of quantization is the axis about which the field rotates, and zero-field states of different m are coupled. In fact, there are two mutually exclusive sets of coupled states, in which $l+m$ is even or odd.

Ionization by a circularly polarized microwave field is easily understood by transforming the problem to a frame rotating with the field.[9] Consider a microwave field in the x-y plane rotating at angular frequency ω about the z axis, the axis of quantization. Transforming the problem to the frame rotating with the field has two effects. First, the field becomes a static field pointing in

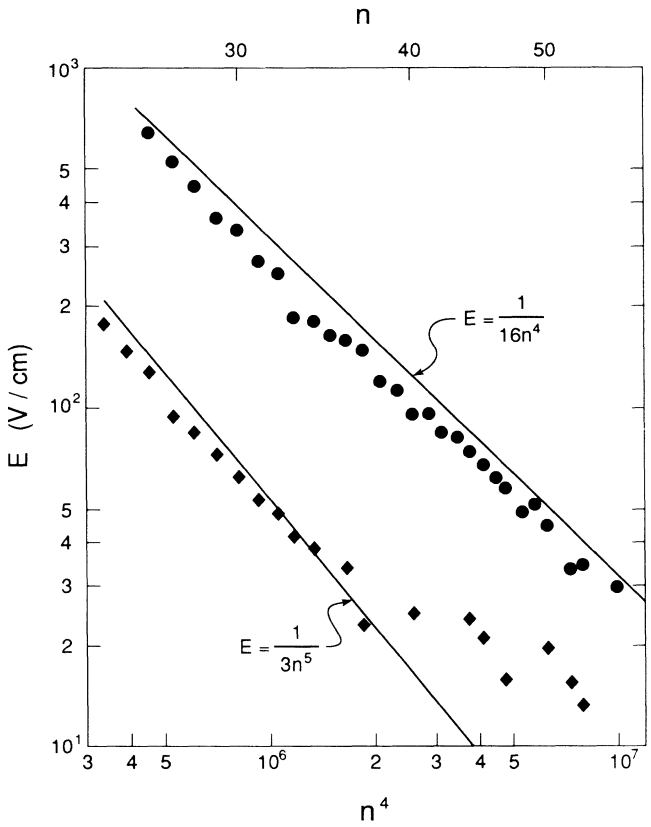

FIG. 3. Ionization threshold fields for linear (■) and circular (●) polarization as a function of n when the atoms are excited in the microwave field.

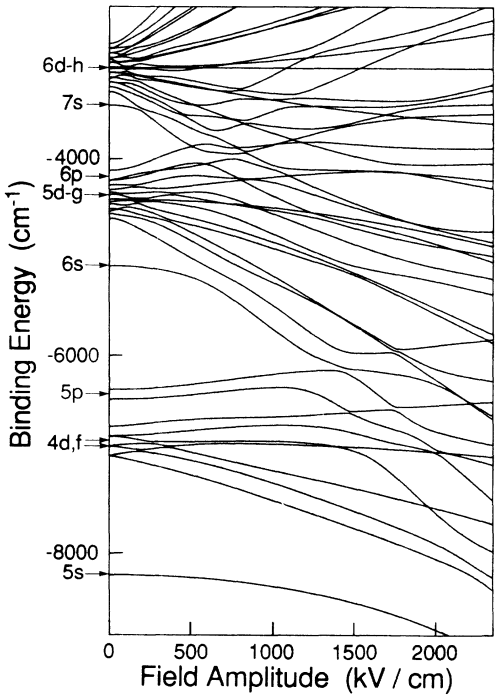

FIG. 4. Energy-level diagram for Na $4 \leq n \leq 6$ and $l+m$ even in a frame rotating at 1500 GHz (50 cm^{-1}), 2% of the $n=4$ to $n=5$ frequency. The energies in the nonrotating frame are shown by the arrows. Note that in zero field $m \neq 0$ levels are displaced from their energies in the nonrotating frame by $\mp m\hbar\omega$. Particularly important is that in strong fields, where the level manifolds of different n overlap, there are pronounced avoided crossings due to the quantum defects of the low-l states, indicating strong couplings, as in a static field. In fields greater than $1/16n^4$, the classical ionization limit, there are similar strong couplings to the Start-induced continuum, and ionization occurs very rapidly.

a fixed direction in the x-y plane. The field does not induce transitions, and the problem is time independent. In the rotating frame there are well defined energy levels, which have been observed[10] and can be found by simply diagonalizing the now time-independent Hamiltonian matrix. First, however, we must take into account the transformation's second effect, that the energies of the zero-field angular momentum states are shifted in energy by $-m\hbar\omega$, a consequence of their properties under rotation.[9] There are too many coupled levels at $n=25$ for us to diagonalize the Hamiltonian matrix. Instead, we have calculated the energy levels in the rotating frame of both H and Na for $4 \leq n \leq 7$, where the number of levels is manageable. In Fig. 4 we show an energy-level diagram as a function of field strength for Na levels of $4 \leq n \leq 6$ and even $l+m$ in a frame rotating at 1500 GHz (50 cm^{-1}), 2% of the $n=4$ to $n=5$ transition frequency. Our frequency, 8.5 GHz, is 2% of the $n=25$ to $n=26$ interval. In Fig. 4 the energies of the states in the nonrotating frame are shown by the arrows at the left-hand side. Note that in zero field m levels are displaced by $\mp m\hbar\omega$ from their energies in the nonrotating frame, as is most apparent for the $5p$ and $6p$ $m = \pm 1$ levels.

If we now turn our attention to high fields at which the manifolds of n and $n+1$ levels overlap, it is clear that the levels do not cross but have pronounced avoided crossings, indicating strong interactions between the levels of different n. If a level lies above the lowest, $m=0$, classical ionization limit, defined by $E=1/16n^4$ (here we ignore any Stark shifts), there are strong couplings to the Stark-induced continuum similar to those producing the avoided crossings of Fig. 4, and ionization occurs rapidly.[11-13] Thus $E=1/16n^4$ is the threshold field for ionization in the rotating frame. In essence, a static field has the same effect whether its coordinate system is rotating or not.

The apparatus is, of course, in the nonrotating frame, and we must convert our understanding to this frame. Fortunately, we excite $|m|=0$ and 1 components which have the same energies, to $\pm\hbar\omega$, in the rotating and nonrotating frames. The energy uncertainty of $\pm\hbar\omega$ is negligible compared to the binding energies of the states in question, and the field-ionization criteria, which come

from the binding energies, are thus the same in both frames, resulting in the prediction of an $E=1/16n^4$ threshold-field dependence, in agreement with our observations.

The calculated H level diagram analogous to Fig. 4 has unobservably small avoided level crossings, indicating vanishing interaction between n states and correspondingly small interactions with the Stark-induced continuum. Thus in hydrogen we would expect the required field to exceed the hydrogenic threshold for ionization, $E=1/9n^4$ (Ref. 4).

The schematic rotating frame energy-level diagram of Fig. 4 also shows why a small amount of ellipticity in the polarization reduces the ionization threshold field. In the rotating frame ellipticity introduces a variation, at frequency ω/π, in the field amplitude, which admits the possibility of Landau-Zener transitions through the avoided crossings to higher, more easily ionized states if $E > 1/3n^5$ (Ref. 4).

In a practical vein, circularly polarized microwaves may provide an m-independent ionization technique and may, as suggested by Molander, Stroud, and Yeazell,[14] be used to produce circular states.

In conclusion, we have shown that the ionization of Rydberg atoms by circularly polarized microwave fields differs dramatically from ionization by a linearly polarized field, as might be expected for a very high-order process.[1,2] What was, at least to us, unexpected is that it can be understood so simply by transforming the problem to a rotating frame and using the notions of static field ionization.

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