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Ion-recoil energy measurement in photoionization of laser-cooled rubidium

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We report a measurement of the ion recoil energy in photoionization of laser-cooled atoms. ^{87}Rb, cooled in a magneto-optical trap, is two-photon ionized by a pulsed laser. The recoil energy is determined by time-of-flight in a weak electric field. A resolution in the range of 1 μeV is demonstrated. Refinement of the technique promises resolution in the range of several neV to be feasible.
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A primary goal in studies of collision-induced atomic reactions involving ionization is the precise resolution of the momenta of all emerging products. This is particularly important, and difficult, in reactions where more than two charged particles appear in the product channel. In this case standard high-resolution spectrometers do not easily lend themselves for coincident detection due to the small acceptance angle. Nevertheless, in specific situations, prudent experimenters have achieved kinematically complete experiments in single and double ionization by electron and photon impact [1–4], at the expense of a reduced detection geometry and long observation times. A solution to these limitations appears through the use of spectrometers with a high angle of acceptance and exploiting the generally miniscule momentum information imparted in the recoil ion. The past decade has seen rapid advances in this field through the combination of position- and time-sensitive detection for atomic particles [5–8]. A recent culmination is the high-resolution ''microscope,'' described by Ullrich *et al.* [9], in which the momentum of a recoil ion is determined in coincidence with other charged products.

A basic limitation in recoil ion experiments is the thermal energy of the system prior to the ionization process. Since recoil energies are typically extremely small, very cold targets are required. These are currently realized in skimmed supersonic jets for which an internal momentum spread of better than ± 0.05 a.u. has been demonstrated, permitting, for example, the measurement of He^+ recoil ions with a resolution $\Delta E_r \approx \pm 30$ μeV [10]. The application of laser-cooled targets in recoil-ion spectroscopy has been discussed in the literature [11] and is expected to permit further refinement in resolution.

Here we report an application of laser cooled atoms in recoil-ion spectroscopy. The experiments were carried out in a magneto-optical trap (MOT) using ^{87}Rb cooled on the $(F=3)^2P_{3/2} \leftrightarrow (F=2)^2S_{1/2}$ transition (see Fig. 1). Recoil ions were formed by two-photon ionization from the excited trapping state using light at 790 nm:

$$\mathrm{Rb}(^2P_{3/2}) + 2h\nu \rightarrow \mathrm{Rb}^+(^1S_0) + e + \Delta E. \qquad (1)$$

In reaction (1) the excess energy ΔE lies at 0.55 eV; the resulting momentum recoil of each product amounts of 0.201 atomic units. The velocity of the recoil ion formed in reaction (1) is 276 cm/s. This velocity is significantly larger than the mean thermal velocity of the laser-cooled atom prior to ionization ($\approx$15 cm/s). Hence the heavy particle trajectory is primarily determined by the recoil momentum. This may be exploited to determine this quantity directly.

Our trap uses three pairs of counterpropagating σ^+ and σ^- laser beams and a magnetic-field gradient of 10 G/cm produced by a pair of anti-Helmholtz coils [12]. The trapping laser system consists of a grating stabilized diode laser with a rms bandwidth of less than 100 kHz (for frequency fluctuations in the range 10 Hz–100 kHz) and an injection-locked diode laser with an output power of about 30 mW in a TEM_{00} mode. A second grating stabilized diode laser, tuned to the $(F=2)^2P_{3/2} \leftrightarrow (F=1)^2S_{1/2}$ transition is used to repump atoms from the nontrapping ground state. A detailed characterization of a similar system can be found in reference [13].

Typically 5×10^6 atoms are trapped from Rb vapor from a solid reservoir at a background pressure of 4×10^{-9} mbar. The trap temperature was determined by an expansion technique [14] to lie in the range of the Doppler temperature (143 μK), corresponding to a momentum spread of Rb atoms of

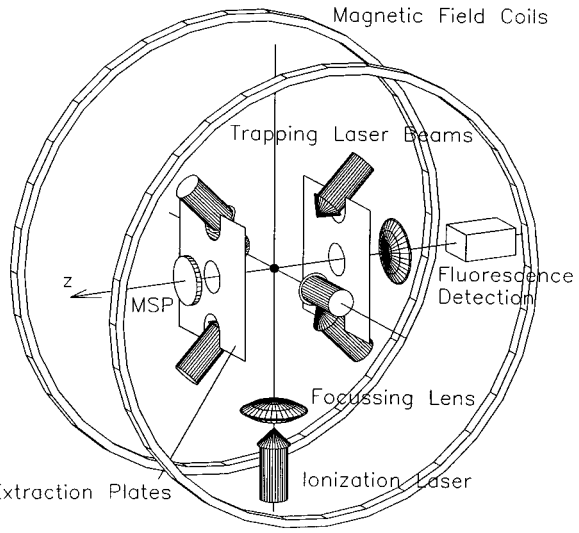

FIG. 1. Schematic of the ion recoil spectrometer with laser-cooled target. Atoms in a standard MOT are ionized by a pulsed laser. The ions are extracted by a weak electric field along the z axis.

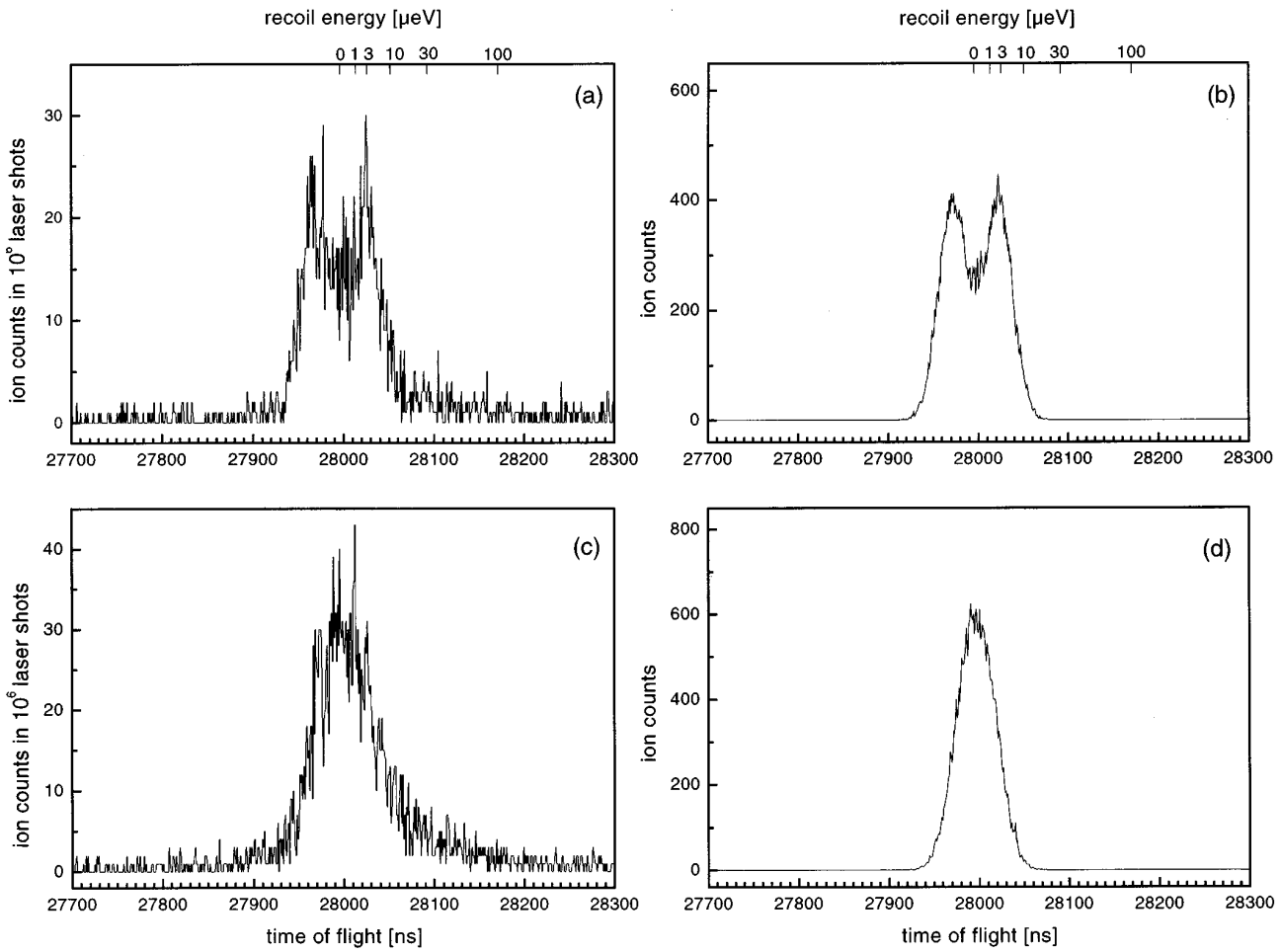

FIG. 2. Experimental time-of-flight spectra of Rb^+ ions with laser polarization (a) parallel and (c) perpendicular to the extraction field. The laser pulse energy was approximately 20 pJ. Monte Carlo simulations of the time-of-flight spectra are shown in (b) and (d) with parameters for (a) and (c), respectively.

<0.01 a.u. While operating the trap continuously, photoionization was achieved with a focused 1-kHz repetition rate Ti:sapphire laser with a pulse duration of 100 fs. In order to avoid space-charge effects on the momentum of the recoil ion, the laser pulse energy was attenuated to below 60 pJ, corresponding to peak intensities in the laser focus of $<10^8$ W/cm^2.

We employed time-of-flight analysis to measure the component of the recoil velocity along the direction of a weak extraction field. The MOT is located inside a 2-in. copper cube with 14 apertures of 12 mm diameter to provide access to the trap region. Two opposing walls of the cube are electrically isolated (shown schematically in Fig. 1), and are kept at ± 2.5 V, respectively. Ions exit the extraction field through an aperture in one end plate and reach a microsphere plate detector after passage of two grids. In simulations using SIMION [15] we find that this electrode geometry establishes an extraction field of 0.77 V/cm in the center of the cube, in agreement with the value determined experimentally from the ion flight time. The arrival time of ions is measured using a digital multichannel analyzer with 500-ps time resolution, triggered by the ionization laser. Typically 1×10^6 shots are accumulated, with the total number of ions recorded falling in the range of 10^3-10^4. The actual number of ions formed is higher, but the secondary emission coefficient of low-energy Rb^+ is known to be aggravatingly low [16]. The two-photon ionization process is expected to lead to ejection of electrons, and hence the recoil ions, preferentially along the laser polarization [17]. For laser polarization parallel to the extraction field, we expect a double-peaked structure in the arrival time spectrum, which arises from the preferential emission of recoil ions along the laser polarization. The time-of-flight difference for reaching the extraction plate from the center of the cube is

$$\Delta t = \frac{2MV_{\mathrm{recoil}}}{e\mathcal{E}_{\mathrm{extr}}}, \tag{2}$$

where M is the ion mass, V_{recoil} the ion recoil speed, and $\mathcal{E}_{\mathrm{extr}}$ the extraction field strength. For our experimental conditions a time-of-flight difference of $\Delta t = 66$ ns is expected.

Typical time-of-flight spectra observed are shown at the left of Fig. 2. For Fig. 2(a) the polarization of the ionization laser was kept parallel to the acceleration field; for Fig. 2(c) it was perpendicular. The spectrum with parallel polarization shows a clearly resolved double-peaked structure, as ex-

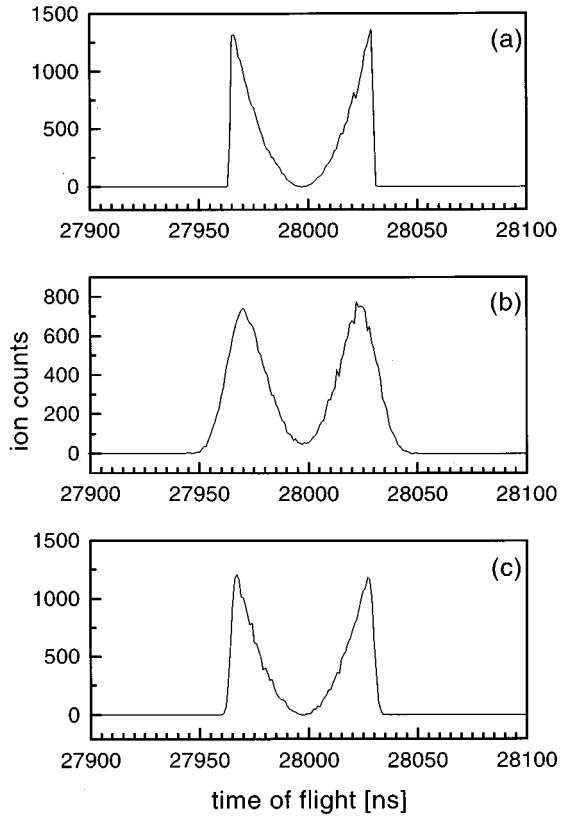

FIG. 3. Monte Carlo simulations of time-of-flight spectra of Rb^+ ions for idealized and realistic experimental parameters: (a) trap temperature 1 μK, laser beam waist 1 μm; (b) trap temperature 1 μK, laser beam waist 20 μm; (c) trap temperature 150 μK, laser beam waist 1 μm.

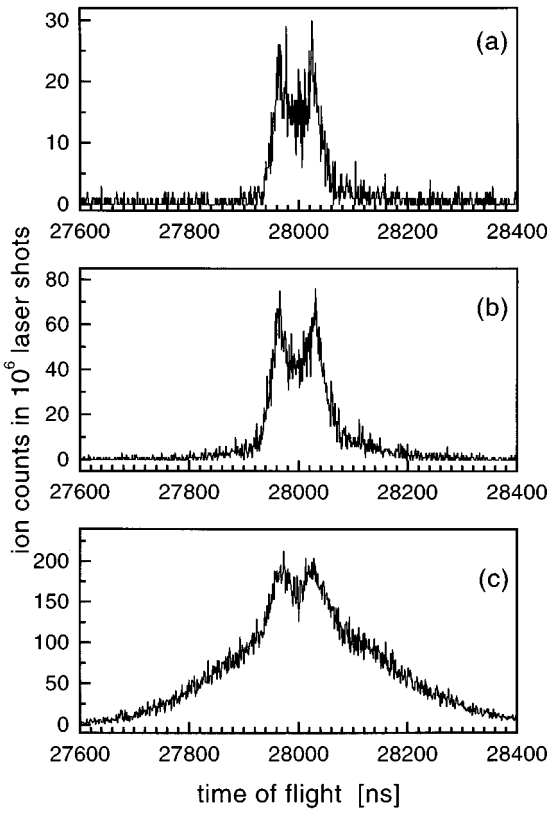

FIG. 4. Experimental time-of-flight spectra for different pulse energies, E_{pulse}, of the ionization laser: (a) $E_{\text{pulse}} \approx 30$ pJ, (b) $E_{\text{pulse}} \approx 20$ pJ, (c) $E_{\text{pulse}} \approx 60$ pJ.

pected. The splitting in the arrival time is in good agreement with the theoretical value predicted. When orienting the polarization perpendicular to the z axis, the arrival time of ions ejected along the direction of laser polarization coincide, leading to a central peak in the spectrum, as may be seen from Fig. 2(c). Monte Carlo simulations of the arrival time spectra expected from reaction (1) under our experimental conditions are shown in Figs. 2(b) and 2(d), respectively. The fitting parameters are the angular distribution of recoil ions and the size of the laser focus (see below). At the top of the figure a recoil energy scale is shown as it appears from Eq. (1). This scale refers to the component of the ion energy falling along the z axis. The recoil energy at which ions are formed in reaction (1) is calculated to be 3.46 μeV.

Several apparatus effects limit the resolution in our current setup. To illustrate the significance of some of these, we next discuss Monte Carlo simulations of ion trajectories under idealized and realistic conditions.

The most significant degradation of resolution arises in the current geometry from the finite volume within which recoil ions are formed: A Gaussian beam waist of 20 μm is expected from our laser beam parameters, but our experimental spectra are more consistent with a waist of 50 μm, indicating that the ionization laser beam profile is not ideal and/or the trapped atom cloud is imperfectly located with respect to the laser focus.

In Fig. 3 simulated arrival time spectra are shown. The angular distribution of photoions is approximated by the differential cross section [17]

$$\frac{d\sigma}{d\Omega} = \frac{\sigma^{\text{iso}}}{4\pi}[1+\beta P_2(\cos\theta)]. \tag{3}$$

Here, σ^{iso} is the total cross section, the angle θ is measured relative to the laser polarization axis, and P_2 is the second Legendre polynomial. A β parameter of 2 was found to best represent the observed spectra.

A 1-μm-diam size source of recoil ions is simulated in Fig. 3(a). The atom temperature is assumed to be 1 μK. In Fig. 3(b) the volume of ion production is taken to be Gaussian with a full width at half maximum of 20 μm under otherwise identical conditions. Substantial broadening appears. A way to eliminate this volume effect is to employ an extraction field in a Wiley-McLaren configuration with spatial focusing [18].

When eliminating the volume effect, the finite temperature of the cold-atom sample should pose the next limitation. The simulation in Fig. 3(c) assumes a Rb temperature of 150 μK, but again for a 1-μm-diam source of ions. Comparison with Fig. 3(a) shows that significant degradation occurs already at this temperature. We are currently attempting to remove the volume effect and employ sub-Doppler cooling techniques [19] to refine our experimental parameters.

Other uncertainties contribute to the smearing of the spectra. Among these is the bandwidth of the ionization laser. The short pulse laser employed here has a measured spectral width of 9 nm and contributes to an uncertainty in recoil

velocity at the level of 4%. Most importantly, residual fields have not yet been actively compensated. Stray fields from electrical feedthroughs and contact potential differences are certainly active, as we find that at extraction fields below 0.4 V/cm ions tend to miss the aperture to the detector. The magnetic field of the atom trap was not found to influence the arrival time spectra at the current level of resolution but will ultimately have to be accounted for, when pushing the resolution into the neV range. A most serious effect can of course arise from space charge: Fig. 4 shows a sequence of spectra obtained at increasing laser pulse energy. Coulomb effects significantly distort the spectra when increasing the average number of ions into the range beyond one ion per shot.

Despite a number of imperfections, which can be overcome in future experiments, we have been able to show that laser-cooled atoms are a versatile tool for high-resolution ion recoil energy spectroscopy, promising access into the range of a few tens neV resolution with standard Doppler-cooling techniques.

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[1] O. Schwarzkopf, B. Krässig, J. Elminger, and V. Schmidt, Phys. Rev. Lett. **70**, 3008 (1993).

[2] A. Huetz, L. Lablanquie, L. Andric, P. Selles, and J. Mazeau, J. Phys. B **27**, L13 (1994).

[3] H. Erhard, K. Jung, G. Knoth, and P. Schlemmer, Z. Phys. D **1**, 1 (1986).

[4] A. Lahmann-Bennani, J. Phys. B **24**, 2401 (1991).

[5] D. P. DeBruijn and J. Los, Rev. Sci. Instrum. **53**, 1020 (1982).

[6] H. Helm, D. P. deBruijn, and J. Los, Phys. Rev. Lett. **53**, 1642 (1984).

[7] R. Moshammer *et al.*, Phys. Rev. Lett. **73**, 3371 (1994).

[8] R. Dörner *et al.*, Phys. Rev. Lett. **72**, 3166 (1994).

[9] J. Ullrich *et al.*, J. Phys. B **30**, 2917 (1997).

[10] V. Mergel *et al.*, Phys. Rev. Lett. **74**, 2200 (1995).

[11] R. Moshammer, M. Unverzagt, W. Schmitt, J. Ullrich, and H. Schmidt-Böcking, Nucl. Instrum. Methods Phys. Res. B **128**, 415 (1997).

[12] E. L. Raab, M. Prentiss, A. Cable, S. Chu, and D. E. Pritchard, Phys. Rev. Lett. **59**, 2631 (1987).

[13] S. Kunze, S. Wolf, and G. Rempe, Opt. Commun. **128**, 269 (1996).

[14] S. Kunze and G. Rempe, in *Proceedings of the Twelfth International Conference on Laser Spectroscopy (TWICOLS) 1995*, edited by M. Allegrini, M. Inguscio, and A. Sasso (World Scientific, Singapore, 1996).

[15] D. A. Dahl, SIMION 3D version 6.0, Princeton Electronic Systems, Inc., 1995.

[16] M. Kaminsky, *Atomic and Ionic Impact Phenomena on Metal Surfaces* (Springer-Verlag, Berlin, 1965).

[17] G. Leuchs and H. Walther, in *Multiphonon Ionization of Atoms*, edited by S. L. Chin and P. Lambropoulos (Academic, New York, 1984), p. 109.

[18] W. C. Wiley and I. H. McLaren, Rev. Sci. Instrum. **26**, 1150 (1955).

[19] P. D. Lett, R. N. Watts, C. I. Westbrook, W. D. Phillips, P. L. Gould, and H. J. Metcalf, Phys. Rev. Lett. **61**, 169 (1988).