Observation of Resolved Zeeman Thresholds in Photodetachment in a Magnetic Field

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Experiments on photodetachment from selenium negative ions in magnetic fields of 6.33 and 7.83 T have allowed the observation of photodetachment from resolved Zeeman levels for the first time. The measured relative detachment strengths for the Zeeman thresholds are not in agreement with the presently available theoretical prediction. Some possible sources of this disagreement are suggested.

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The problems of atomic structure and collisions in strong magnetic fields have been the subjects of numerous theoretical and experimental investigations.[1] They are fundamental, unsolved problems in atomic physics and are of considerable interest for astrophysics because of the very large magnetic fields which exist near some stars.[2] Despite this interest, the only experimental information currently available on continuum states of electron plus neutral atomic systems in a magnetic field arises from a few experiments involving photodetachment from atomic and molecular negative ions.[3-5] Such experiments may be of substantial practical interest as well since they suggest the feasibility of highly efficient and possibly state-selective photoneutralization of fast negative-ion beams.[6] In these experiments the data were found to be consistent with the description of Blumberg, Itano, and Larson[7] (subsequently referred to as BIL) which made major simplifying assumptions including the neglect of a final-state interaction. This theoretical description has been challenged,[8] expanded,[4] and discussed[9-12] but, perhaps despite its simplicity, it has been consistent with available data. The situation suggests that further real progress must rely upon experiments which go significantly beyond those previously performed.

All of the experiments on photodetachment in a magnetic field to date have involved detachment from unresolved Zeeman levels since the splittings provided by the magnetic field were less than the motional broadening. We report here the results of new experiments done with motional broadening somewhat smaller than that in the earlier experiments and with much larger Zeeman splittings that allow the observation of photodetachment from individual Zeeman levels. The resolution of magnetic structure has been increased by an order of magnitude over that achieved in earlier experiments. The present measurements were carried out in a new Penning ion-trap apparatus which uses a superconducting solenoid to provide fields of up to 8 T. The major finding of these new experiments is that photodetachment from the resolved Zeeman sublevels shows a clear departure from the relative strengths predicted by BIL.

The new Penning trap is of the standard design[13-15] with ring and end-cap electrodes in the approximate shape of truncated hyperboloids. The ring and end-cap supports are made from oxygen-free high-conductivity copper and the end caps themselves are made from gold screens. Compensation electrodes are provided between the ring and end caps.[15] The trap was mounted inside a 3.8-cm-diam stainless-steel vacuum pipe which is inserted in the room-temperature bore of a superconducting solenoid. The solenoid provides magnetic fields of up to 8 T and has superconducting shim coils to reduce relative inhomogeneities in the fields to about 10^{-6} in a 1-cm^3 volume. An electron gun placed just below the solenoid and to the side of the axis of the trap produces the low-energy electrons used to create the ions in the trap. Photodetachment was done with a laser beam which was directed through the trap along the magnetic field. The strength of the magnetic field was measured by observation of the cyclotron frequencies of the trapped ions.

The experimental cycle is similar to that used in previous experiments.[3-5] The Se^- ions were created by dissociative attachment of low-energy electrons to CSe_2 which was leaked into the vacuum system at a pressure in the vicinity of 10^{-7} Torr. After the electron beam was turned off, the number of ions in the trap was measured by driving of the ions' axial motion and observation of the currents induced on the ring electrode. The ions were then illuminated by light from a dye laser tuned near the photodetachment threshold at 613 nm while the amount of light passing through the trap was monitored by a photodiode. The laser frequency was monitored with a combination of a wavemeter, a Fabry-Perot spectrum analyzer, and an iodine absorption cell. When the integrated light power reached a predetermined level the laser beam was blocked and the number of ions remaining in the trap was measured. The whole cycle of filling the trap, measuring the ion number, detaching some fraction of the ions, measuring the number again, and emptying the trap took on the order of 15 s. Several such cycles were averaged to produce one datum point.

Photodetachment data were collected at two magnetic fields, 6.33 and 7.83 T, with a variety of different conditions of light intensity, light polarization, and CSe_2 pressure. All of these quantities were held constant in a given run while the laser frequency was scanned. The thresholds studied involved transitions from the $^2P_{3/2}$ state of the negative ion to the $^3P_2$ state of the neutral. The closest spaced Zeeman components of this threshold lie at energies differing by one-sixth of $\mu_0 B$, where μ_0 is the Bohr magneton. Twenty-two Zeeman thresholds are possible for each Landau level of the detached electron, occurring over a range of $4\mu_0 B$. The thresholds repeat for each new Landau level. Since the photodetachment thresholds for detachment to the lowest Landau level ($n=0$) are spread over a range of more than 350 GHz and since the data points were spaced by as little as 1.5 GHz in order to observe fully the structure in the cross section, laser-frequency scans could generally be taken only over a fraction of the total range while allowing sufficient time for averaging of noise and monitoring of drifts in a single run. The lack of significant drift in the signal was verified by periodic measurement of the detachment signal a few hundred gigahertz below the threshold. Each part of the $n=0$ spectrum was examined in some detail. The data are presented as the fraction of ions surviving photodetachment plotted as a function of laser frequency.

It is useful to consider first the results predicted by BIL for conditions similar to those realized in the experiments in order to understand the experimental data. The predicted ion survival ratio for light polarization and intensity comparable to that used for some of the data is shown for the whole $n=0$ region at 6.33 T in Fig. 1. The prediction is done with a small fraction of π-polarized light. The presence of such light, apparently due to scattering off the gold mesh end cap, was observed experimentally. In this prediction, 85% of the σ-polarized light carries angular momentum, q, of $+1$ so that the highest ($\Delta m = q = +1$) thresholds are accentuated. Resolved Zeeman thresholds are readily apparent as rapid changes in ion number with laser frequency. This prediction used an ion temperature of 670 K to produce motional broadening similar to that observed in actual data. The ion temperature is lower than the temperatures in the range of 900 to 1200 K obtained in earlier experiments. The difference is probably due to separation of the hot filament from the trap and perhaps to the higher background gas (CSe_2) pressures used in this experiment. Motional broadening of the features in the photodetachment cross section results from a combination of Doppler and motional Stark shifts.[7] The lower temperatures and higher mass of Se^- imply motional shifts which are a factor of about 1.8 smaller than those present in the earlier experiments on S^-. More significantly, the higher fields used in the present experiments provide Zeeman splittings which are a factor of 5 larger. The combined effect of these two factors is an increase in resolution of the thresholds by a factor of about 9. In the present experiment the combined motional shifts are less than 1 GHz and the Zeeman thresholds are separated by over 18 GHz (at 7.83 T). The comparison of these two numbers suggests resolution somewhat better than that actually obtained, however, since the motionally broadened cross section appears as an asymmetric peak a few gigahertz wide. No attempt was made in the present experiment or analysis to account for isotopic shifts in the photodetachment thresholds. The mass resolution of the detection scheme was such that a few isotopes would contribute to the signal, but since the isotopic shifts in the photodetachment threshold should be on the order of 1 GHz or less, the isotopic differences should not have a significant effect on the shape of the observed structure.

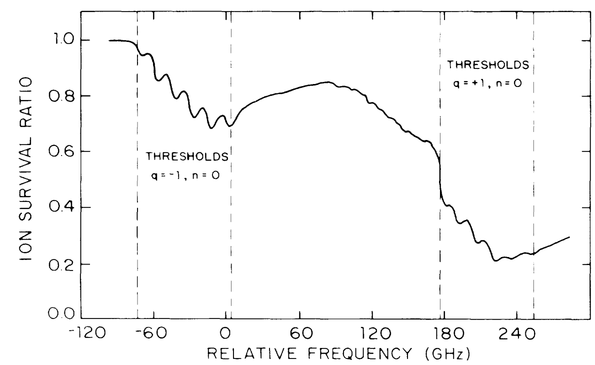

FIG. 1. A theoretical prediction of ion survival ratios as a function of laser frequency that would be expected for a particular light geometry and a magnetic field of 6.33 T. The prediction is based on the description of Ref. 7. Most of the light has σ polarization, and 85% of that light carries angular momentum (q) of $+1$ so that the highest thresholds shown are accentuated. The sets of thresholds for different cyclotron states (n) of the photodetached electron are well separated. Individual Zeeman thresholds also can be readily seen as rapid changes in the ion number with laser frequency. The frequency zero corresponds to the zero-magnetic-field photodetachment threshold.

Figure 1 shows six resolved Zeeman thresholds in the $q=-1$, $n=0$ region of the spectrum. Another threshold, involving the $m_J = -\frac{3}{2}$ state of the negative ion, lies just below the lowest-frequency threshold apparent, but it cannot be seen in the figure since BIL predicts zero strength for this threshold. A little structure can be seen in the region between the $q=-1$ and $q=+1$, $n=0$ thresholds, but the most obvious effect of the $q=0$, $n=0$ and $q=-1$, $n=1$ thresholds which lie in this region is a fairly smooth reduction of the ion number in a region where the cross section due to the $q=-1$, $n=0$ thresholds is decreasing with increasing light frequency. The first Zeeman threshold in the $q=+1$ part of the spectrum results in a large change in the ion number since this threshold involves the first substantial detachment from the $m_J = -\frac{3}{2}$ state.

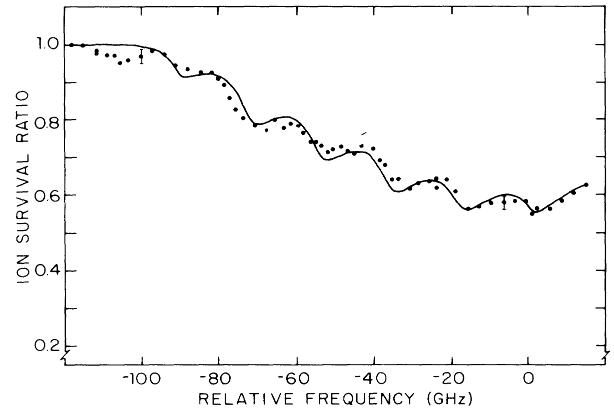

FIG. 2. Experimental data points for the $n=0$, $q=-1$ thresholds at 7.83 T fitted by the theory illustrated in Fig. 1. The lack of agreement in the region at and below the first of the predicted thresholds is reproduced consistently in all of the data.

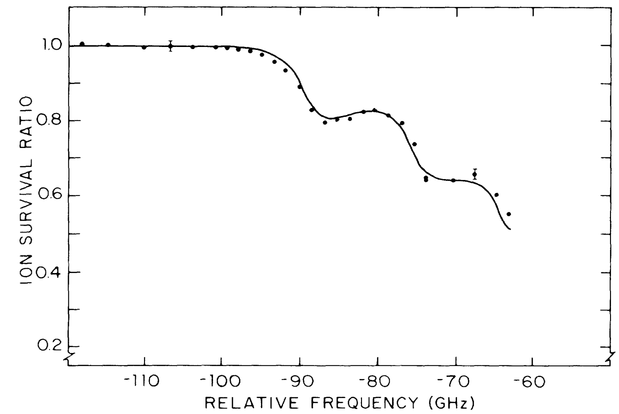

FIG. 3. Experimental data points for the first three observed $n=0$, $q=-1$ thresholds at 6.33 T together with a fit by the theory illustrated in Fig. 1 but with the experimental strengths presented in Table I. The thresholds appear at different positions than in Fig. 2 because of the difference in the strength of the magnetic field. The threshold clearly visible near −90 GHz is predicted in Ref. 7 to have zero strength.

An example of experimental data for the $q=-1$, $n=0$ thresholds at 7.83 T is shown in Fig. 2. The data points are shown with a least-squares fit by the prediction of BIL. Zero frequency (in all of the figures) is at the position of the zero-magnetic-field detachment threshold. The signal-to-noise ratio, typical for much of the data, is sufficient to see the resolved Zeeman thresholds and to see a deviation from the prediction, especially in the region of the lowest-frequency thresholds. The first threshold, involving the $m_J=-\frac{3}{2}$ state of the ion, is present with nonzero strength. Also, the second observed threshold appears to have smaller strength than predicted. Both of these observations are supported by all of the data collected in this region, at both 6.33 and 7.83 T. The clearest evidence for these conclusions obtained in a single run is shown in Fig. 3, where just the first three thresholds have been probed. These data were taken at a field of 6.33 T and fitted by a cross section with a shape given by BIL but with strengths for the Zeeman thresholds derived from several experimental runs as discussed below. The threshold evident near a frequency of −90 GHz is the one predicted to have zero strength. (This threshold is at a different position than in Fig. 2 because of the different magnetic field.) The fit demonstrates that the shape of the predicted cross section appears to be reasonable, but that the strengths are not. The data shown in Fig. 3 were taken with high pressures and long light intervals, conditions which produced the best signal-to-noise ratio, but which also produced evidence for collisional redistribution of Zeeman populations which can affect measurements of relative strengths of the Zeeman thresholds. Little effect of collisions was observed on the first two thresholds, however, and in any case, collisional redistribution of populations could not change the apparent strength of a threshold if it in fact had zero strength to begin with.

The experimental strengths for four thresholds derived from fits to data taken with relatively low pressures and short times are presented in Table I along with the strengths predicted by BIL. The experimental strengths are derived from seven runs including the one shown in Fig. 2. The strengths of all four thresholds appear to deviate from the theoretical strengths. The $m_J=-\frac{3}{2}$ threshold is merely the most dramatic. The strengths are obtained on the assumption that the four Zeeman levels of the negative ion are initially present with equal populations, and the sum is normalized to that of the theoretical prediction. In principle, on the basis of the geometry of the electron beam and the field, the ions could be formed with unequal Zeeman populations. It seems unlikely, however, that significant alignment could be produced in the dissociative attachment process. Such concerns appear to be put to rest by the experimental strengths themselves which suggest that neither align-

TABLE I. Experimental and theoretical (Ref. 7) strengths of the first four $\Delta m=-1$ Zeeman components in the $Se^-(^2P_{3/2})$ to $Se(^3P_2)$ photodetachment threshold. The experimental strengths were found by a least-squares fit to seven sets of data with use of the cross section of Ref. 7 with variable strengths for these thresholds.

Transition $m_J(Se^-)$	$m_J(Se)$	Theoretical strength	Experimental strength
$-\frac{3}{2}$	−2	0	0.16(6)
$-\frac{1}{2}$	−1	0.5	0.34(12)
$\frac{1}{2}$	0	1.33	1.53(26)
$\frac{3}{2}$	1	1.5	1.30(32)

ment nor polarization in the initial state could be responsible for the observed deviations from the prediction. Of course, for the first threshold a consideration similar to that relevant to collisional redistribution applies. No distribution of initial Zeeman populations could cause the appearance of the first $m_J = -\frac{3}{2}$ threshold if its strength were truly zero.

While the data presented here do not suggest that the shape of the cross section predicted by BIL is incorrect, they present a clear disagreement with the relative sizes of the cross sections at the resolved Zeeman thresholds. Within the context of the BIL description the prediction of the relative sizes is based upon some obvious assumptions. For example, it is assumed that the effect of the magnetic field on the continuum wave function in the region near the atom is small. This means that the angular part of the bound-continuum matrix element can be treated just as it would be if there were no field present and that the detachment cross section is dominated by transitions to an s-wave continuum. Also, it is assumed that the angular matrix element can be calculated based on LS coupling in the atom and negative ion. In addition, the effects of any final-state interaction are explicitly neglected.[4] The failure of these assumptions can lead to a nonzero strength for the first Zeeman threshold and an alteration of the strengths for the other thresholds. Very simple estimates suggest that it is unlikely that the failure could be due to the effect of the magnetic field on the continuum wave function, but in any case, the present data make it clear that further experiments and calculations are needed to understand more fully the process of photodetachment in a magnetic field.

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[1]For recent reviews see, M. R. C. McDowell and M. Zarcone, Adv. Atom. Mol. Phys. **21**, 255 (1985); J. C. Gay, in *Progress in Atomic Spectroscopy, Part C,* edited by H. J. Byer and H. Kleinpoppen (Plenum, New York, 1984), p. 177; C. W. Clark, K. T. Lu, and A. F. Starace, *ibid.*, p. 247; D. Kleppner, M. G. Littman, and M. L. Zimmerman, in *Rydberg States of Atoms and Molecules,* edited by R. F. Stebbings and R. B. Dunning (Cambridge Univ. Press, Cambridge, England, 1983), p. 31.

[2]H. Ruder, H. Herold, W. Rosner, and G. Wunner, Physica (Amsterdam) **127B**, 11 (1984).

[3]W. A. M. Blumberg, R. M. Jopson, and D. J. Larson, Phys. Rev. Lett. **40**, 1320 (1978).

[4]D. J. Larson and R. C. Stoneman, Phys. Rev. A **31**, 2210 (1985).

[5]R. C. Stoneman and D. J. Larson, J. Phys. B **19**, L405 (1986).

[6]A. I. Herschovitch and E. A. Hinds, in *Production and Neutralization of Negative Ions and Beams—1983,* edited by K. Prelec, AIP Conference Proceedings No. 111 (American Institute of Physics, New York, 1984), p. 763.

[7]W. A. M. Blumberg, W. M. Itano, and D. J. Larson, Phys. Rev. A **19**, 139 (1979).

[8]C. W. Clark, Phys. Rev. A **28**, 83 (1983).

[9]W. P. Reinhardt, J. Phys. B **16**, L635 (1983).

[10]W. P. Reinhardt, in *Atomic Excitation and Recombination in External Fields,* edited by M. H. Nayfeh and C. W. Clark (Gordon and Breach, New York, 1984), p. 85; C. W. Clark, *ibid.*, p. 17.

[11]O. H. Crawford and D. A. Micha, Bull. Am. Phys. Soc. **30**, 856 (1985).

[12]See McDowell and Zarcone, Ref. 1.

[13]H. G. Dehmelt, Adv. Atom. Mol. Phys. **3**, 53 (1967), and **5**, 109 (1969).

[14]D. J. Wineland, W. M. Itano, and R. S. Van Dyck, Jr., Adv. Atom. Mol. Phys. **19**, 135 (1983).

[15]L. S. Brown and G. Gabrielse, Rev. Mod. Phys. **58**, 233 (1986).