Magnetic trapping of NH molecules with 20 s lifetimes

E Tsikata[1,2,5]**, W C Campbell**[1,3]**, M T Hummon**[1,2]**, H-I Lu**[1,4]
and J M Doyle[1,2]

[1] Department of Physics, Harvard University, Cambridge, MA, USA
[2] Harvard/MIT Center for Ultracold Atoms, Cambridge, MA, USA
[3] University of Maryland, Joint Quantum Institute, College Park,
MD 20742, USA
[4] School of Engineering and Applied Sciences, Harvard University,
Cambridge, MA, USA
E-mail: tsikata@fas.harvard.edu

Abstract. Buffer gas cooling is used to trap NH molecules with $1/e$ lifetimes exceeding 20 s. Helium vapor generated by laser desorption of a helium film is employed to thermalize 10^5 molecules at a temperature of 500 mK in a 3.9 T magnetic trap. Long molecule trapping times are attained through rapid pumpout of residual buffer gas. Molecules experience a helium background gas density below $1 \times 10^{12}\,\mathrm{cm}^{-3}$.

Contents

[5] Author to whom any correspondence should be addressed.

IOP Institute of Physics **Φ** DEUTSCHE PHYSIKALISCHE GESELLSCHAFT

1. Introduction

Avid interest in cold molecules arises from their potential utility in diverse areas of physics. Cold molecules are employed in studies of the temporal variation of fundamental constants [1, 2] and in searches for a permanent electric dipole moment of the electron [3, 4]. Infrared spectroscopy of molecules provides important information about their abundance in stellar environments like cold dark clouds [5]–[7] and can thus give insight into the evolution of these environments. The strong electric dipole–dipole interaction between molecules polarized by external electrical fields may lead to the creation of condensed matter systems with novel properties [8] and the use of molecular ensembles as bits in quantum computers [9, 10].

In many experiments, the trapping of molecules facilitates their study. Trapped molecular samples are useful in understanding spin depolarization and collisional quenching [11, 12]. The spontaneous radiative decay of molecules in rovibrationally excited levels is directly and most precisely measured by observing the temporal evolution of excited state populations [12, 13]. These transitions, typically having lifetimes of several milliseconds, can be monitored when dilute samples of molecules are confined in free space using electromagnetic fields. Long trapping times (compared to the spontaneous lifetime) permit more accurate extraction of state lifetimes. Techniques to increase molecular phase space densities through collisions also benefit from increased confinement times, as efficient cooling requires sufficient time for molecule populations to thermalize to the temperature of the refrigerant (in sympathetic cooling) [14, 15] or to rethermalize (evaporative cooling) [16]–[18]. Trap lifetimes of around ten seconds were required to evaporate the first atomic samples to quantum degeneracy [19].

This paper reports the use of a variant of the buffer gas technique to cool and magnetically trap NH molecules with $1/e$ lifetimes exceeding 20 s. We have previously reported the use of the buffer gas technique to study the spin relaxation of NH molecules in collisions with helium [11]. In the trapping conditions of that experiment ($T = 710$ mK, $B_{max} = 3.9$ T, $n_{He} \sim 5 \times 10^{14}$ cm^{-3}, where T is the temperature, B_{max} is the depth of the magnetic trap, μ is the magnetic moment of the molecule and n_{He} is the helium density), molecule trap lifetimes were limited to around 200 ms by inelastic buffer gas collisions. The dimensionless parameter η, given by $\eta = \mu B_{max}/k_B T$ ($\eta = 7.5$, for the conditions described above) is an important figure of merit in trapping experiments, expressing the ratio of the thermal energy of the particles to the depth of the confining potential. Enhanced molecule lifetimes in the current experiment (at a value of η similar to that in [11]) are achieved through a 1000 fold reduction in the helium background density on a timescale shorter than the trapping time.

To dissipate the translational and rotational energy of the molecules we produce from our room temperature source, we require helium densities of approximately 1×10^{15} cm^{-3}. At these densities, the molecule trap lifetime is determined by the competing effects of diffusion and spin depolarization. Diffusion enhancement of the trap lifetime is proportional to n_{He}; spin depolarization produces loss that reduces the trap lifetime as $1/n_{He}$. As the buffer gas density is reduced from the densities required for loading our magnetic trap, diffusion and spin depolarization become less significant and the effect of trap evaporation (expulsion of molecules due to elastic collisions with the buffer gas) dominates. Trap evaporation has a roughly exponential dependence on both the trapping parameter η and the helium density. Longer trapping durations may thus be achieved by reducing the ambient helium density, trapping colder molecules and employing deeper magnetic traps. Previous work [20] has shown
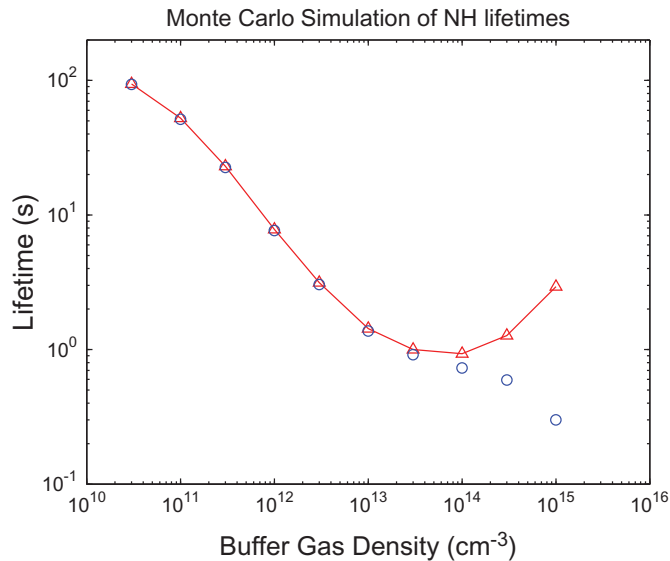
IOP Institute of Physics Φ DEUTSCHE PHYSIKALISCHE GESELLSCHAFT

Figure 1. Monte Carlo simulation of trapped molecule lifetimes in the presence of helium background gas. The molecule and buffer gas temperatures are assumed to be 0.5 K. The red triangles show the effect of evaporative loss, arising from elastic collisions with the buffer gas, on the molecule lifetime. The blue circles indicate the observed lifetime when the effects of both spin depolarization (inelastic loss) and trap evaporation are included.

that reductions in the helium density must occur rapidly compared to the molecule trap lifetime for a significant number to be retained in low background gas conditions.

Trap evaporation is studied by performing a Monte Carlo simulation of molecule trajectories [21, 22]. Figure 1 presents the results of the simulation of NH molecules embedded in a helium vapor background. Collisions are modeled as hard sphere scattering events. The buffer gas temperature is assumed to be 500 mK, the cell temperature. The elastic collision cross-section between NH and ^{3}He is $2.7 \times 10^{-14} \, cm^{-2}$ [11]. Red triangles show the effect of elastic collisions between molecules and buffer gas atoms. In the absence of inelastic collisions, a minimum lifetime is predicted to occur at a density of approximately $10^{14} \, cm^{-3}$, when the background gas is too dilute to enforce diffusive motion but frequent collisions can eject molecules from the trap. The spin depolarization rate for NH–^{3}He collisions, k_{in} (studied in [11]), was found to be $3.0 \pm 0.9 \times 10^{-14} \, cm^3 \, s^{-1}$. The blue circles result from including the effects of spin-depolarizing collisions on the molecule lifetime.

2. Experimental apparatus and technique

The experimental apparatus employed to trap molecules for longer durations, figure 2, is similar to that described in [11]. NH molecules, produced in the form of a supersonic beam by glow discharge of ammonia, are injected into a copper cell housed in the bore of a superconducting magnet (anti-Helmholtz configuration). The cell is thermally anchored to a ^{3}He refrigerator and sits at approximately 500 mK. In the previous experiments, the molecules thermalized to
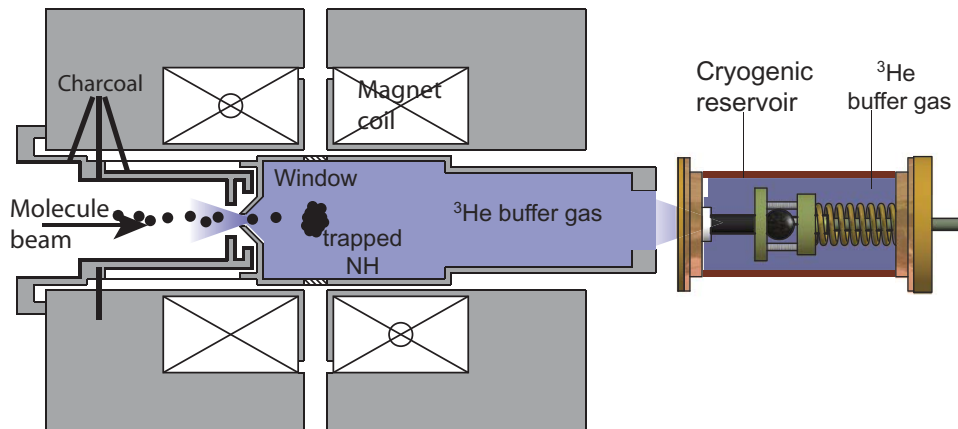
Figure 2. Schematic drawing of the experimental apparatus. The molecule beam propagates through the vacuum space to enter the cell, housed in the bore of the trapping magnet. Buffer gas, supplied by the pulsed reservoir, fills the cell and allows the molecules to thermalize to the cell temperature. The molecules are detected with laser light that passes through window ports in the magnet midplane.

the cell temperature through collisions with helium supplied by steady flow through a fill line attached to the cell. The fixed flux into the cell through the fill line and out of the cell through the 1 cm diameter front aperture established a constant helium density within the cell. In the current apparatus, a rapid reduction in the helium density after loading is attained by creating an additional 3.80 cm diameter aperture at the back end of the cell. A pulse of helium gas, provided by an external reservoir physically disconnected from the trapping cell, is used to thermalize the injected molecules in lieu of the steady helium flow.

The helium reservoir is a copper chamber with a fill line for gas supply and an aperture that permits the gas to be delivered to the cell. The reservoir volume is $200\,\text{cm}^3$; the aperture diameter is 0.64 cm. A phosphor bronze spring presses a PTFE disc against the bottom reservoir flange to seal the aperture. Thermal connection to the ^{3}He refrigerator fixes the reservoir temperature at 900 mK.

The molecule loading procedure is as follows. The reservoir seal is opened and helium fills the cell at an approximate density of $10^{15}\,\text{cm}^{-3}$ for 20 ms. The molecule beam is fired into the helium vapor, which flows out of the cell and into the vacuum space. The molecules persist in the trap with $1/e$ lifetimes of a few seconds (figure 3), limited by the slow desorption of helium from a film bound to the cell surface. Molecules are detected by collecting fluorescence from the spontaneous decay of the $|A\,^3\Pi_2, v'=0, N'=1\rangle$ state, after laser excitation of the $|X\,^3\Sigma^-, v''=0, N''=0\rangle \rightarrow |A\,^3\Pi_2, v'=0, N'=1\rangle$ transition.

The lifetime limit imposed by film desorption may be circumvented by a modification of the loading procedure. The reservoir is again used to fill the cell with helium. The film is then allowed to thin over time. 45 s after the actuation of the reservoir, 4 mJ of energy from a 532 nm YAG laser is incident on the cell; the YAG pulse elevates the cell temperature from 520 to 590 mK and desorbs helium from the cell walls. The molecular beam is timed to fire with the YAG pulse, and the desorbing helium thermalizes a portion of the molecules, which
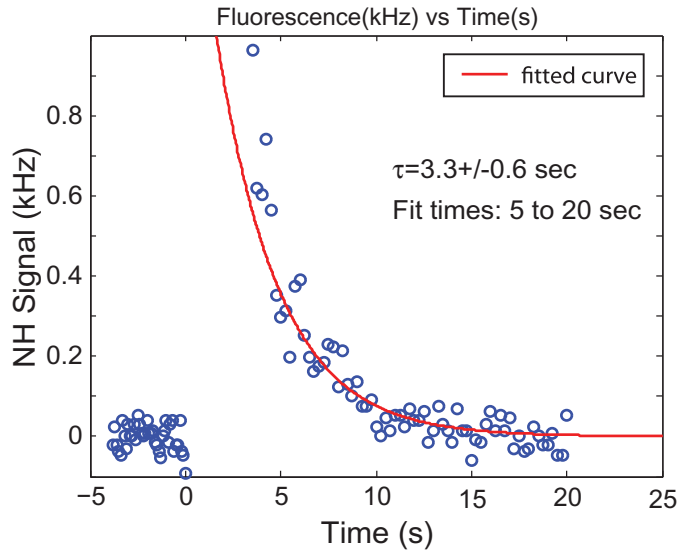
Figure 3. Decay of molecule fluorescence after loading with the buffer gas reservoir. Points before 0 s show the detection background.

are retained in the trap. From the measured lifetime and the Monte Carlo simulation, we infer a background helium density between 10^{11} and 10^{12} cm^{-3} in the trapping region. The loading procedure permits smaller numbers of molecules (a factor of 1000 less) to be trapped for long durations. The extension of this technique to thermalize molecules delivered as a slow beam might lead to an optimal situation whereby substantial numbers of molecules are retained for long periods of time.

Figure 4(a) shows the temporal decay of the molecular fluorescence. Points before 0 s show the background detection level, principally set by dark counts of photomultiplier detector. The fluorescence trace is fit to a single exponential, with a $1/e$ lifetime of 24.6 ± 6.9 s. Data beyond 10 s are used to perform the fit to minimize the effect of cell cooling on the measured lifetime. Trapped molecule numbers are estimated to be 10^5 at a peak density of 10^5 cm^{-3}. The inset (figure 4(b)) shows fluorescence spectra at different times. The red trace shows a spectrum recorded 150 ms after the firing of the molecular beam. The blue circles are a spectrum measured 10 s after the molecular beam launch. The fluorescence signal for the early time spectrum is reduced by a factor of 300 to permit comparison to the spectrum at longer time. The spectra are obtained by sweeping the probe laser frequency and counting the photons emitted by spontaneous decay of electronic excited state ($|A\,^3\Pi_2,\ v'=0,\ N'=1\rangle$). The peaks arise from coincidence between the laser frequency and the frequency of Zeeman transitions between the ground and excited states.

Left peak, $\Delta m_J = 0$: $|X\,^3\Sigma^-,\ v''=0,\ N''=0,\ m_J=1\rangle \rightarrow |A\,^3\Pi_2,\ v'=0,\ N'=1,\ m_J=1\rangle$.

Right peak, $\Delta m_J = 1$: $|X\,^3\Sigma^-,\ v''=0,\ N''=0,\ m_J=1\rangle \rightarrow |A\,^3\Pi_2,\ v'=0,\ N'=1,\ m_J=2\rangle$.

In the inhomogeneous trapping field, the peaks are broadened by the differing g-factors of the Zeeman sublevels. The $\Delta m_J = 0$ transition is thus 1.5 times the width of the $\Delta m_J = 1$ transition (cf [23] for calculation of magnetic moments of Hund's case (b) molecules). Optical
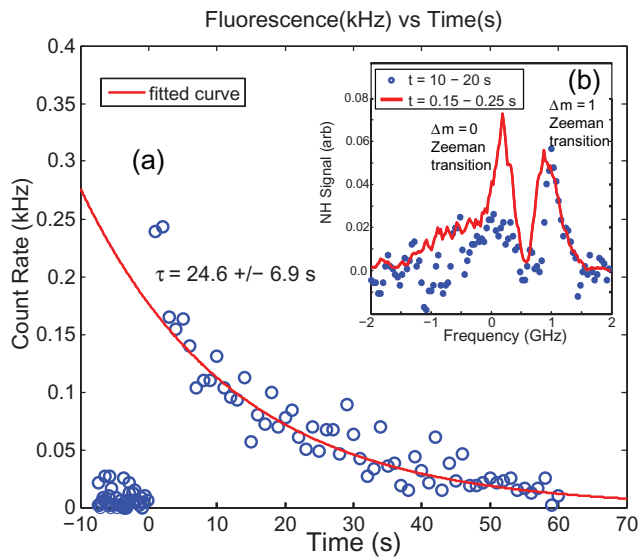
Figure 4. (a) Temporal decay of molecule fluorescence. The fit is performed after 10 s to reduce the effect of cell cooling on the lifetime. (b) NH spectra taken at different time intervals. The spectrum recorded between 0.15 and 0.25 s after the triggering of the molecular beam is given by the red trace. The spectrum recorded between 10 and 20 s is shown by the blue circles. The fluorescence intensities of the two spectra are scaled for comparison—the signal at 10 s is 300 times weaker than at 0.15 s. Consult the text for the explanation of the difference in the relative heights of the spectral features.

pumping leads to the difference in the relative peak heights of the spectra at different times. To record long trapping lifetimes, we excite the molecules by modulating a low-intensity laser beam at a frequency of 1 MHz for 10 ms of each second (to prevent rapid optical pumping of the molecule population to untrapped states). Molecules excited to the $|A\,^3\Pi, m_J = 2\rangle$ state ($\Delta m_J = 1$ transition) are more likely to decay to a trapped state ($|X\,^3\Sigma^-, m_J = 1\rangle$) and be recycled than molecules excited to the $|A\,^3\Pi, m_J = 1\rangle$ state ($\Delta m_J = 0$ transition). To obtain the short time spectrum, the probe laser interrogates the molecules continuously. The laser intensity is sufficient to cause optical pumping, and the fluorescence peaks are of comparable intensity.

Optical pumping by blackbody radiation has been shown to limit trapping times in room temperature experiments [24]. Hoekstra *et al* calculate maximal room temperature trapping lifetime for NH to be under 10 s. At the 1 K temperature of our experiment, the blackbody pumping rate is strongly suppressed (by a factor of $\sim 10^{10}$) and plays no part in determining the molecule lifetime.

3. Conclusions

The trapping of molecules in conditions of lower background gas density increases the prospects of success for a number of collisional experiments. Recent experiments have obtained absolute cross-section measurements for magnetically trapped OH molecules colliding with

helium and molecular deuterium beams. In these studies, the OH lifetime was limited to 432 ms by collisions with background gas at a pressure of 7.5×10^{-9} torr [25]. Our group has previously demonstrated the simultaneous loading and magnetic trapping of nitrogen atoms and NH molecules [26]. The interaction of these species in an environment with a low density of helium will facilitate more sensitive study of the collisional properties of this system. Favorable collisional properties (low inelastic loss rates) could lead to nitrogen being used as a refrigerant for the NH molecules, enabling the sympathetic cooling of the molecule. We also expect increased loading efficiency if the molecules are supplied to the trapping region as a 4 K effusive beam. We have developed such beams [27, 28] with a variety of species. Higher densities of accumulated molecules and long trapping times could allow us to study low-temperature molecule–molecule interactions. While our molecule temperatures currently exceed those attained using other methods (such as Stark deceleration techniques), additional cooling steps may permit dramatic temperature reductions and increases in molecular phase space densities [29].

Long trapping times in our experiment might also permit us to perform high precision measurements of the lifetimes of the metastable states of radical species. In our measurement of the radiative lifetime of the first vibrational level of NH, the 37 ms lifetimes was a significant fraction of the 200 ms trapping time; extraction of the radiative lifetime required correction for the effect of a finite trapping time. In the conditions of the current experiment, the radiative lifetime is less than one per cent of the molecule trapping time. The reduction in the ambient helium density also minimizes the uncertainties associated with the quenching of the excited vibrational state (our dominant systematic uncertainty) and facilitates the measurement of longer lifetimes for metastable states of other trappable species.

The low helium background present in the experiment also suggests that this variant of buffer gas loading can be applied to thermalize and trap particles with unfavorable ratios ($\gamma = \sigma_{\mathrm{elastic}}/\sigma_{\mathrm{inelastic}}$) of elastic to inelastic cross-sections, potentially extending the number of species that can be studied. High rates of inelastic loss due to collisions with helium limited the trapping lifetimes of the highly paramagnetic molecules chromium hydride ($\gamma \sim 9000$) and manganese hydride ($\gamma \sim 500$) to 100 ms [30]. Employing a transient burst of helium to thermalize the molecules and then rapidly removing the residual gas could lead to trap lifetimes of several tens of seconds for these molecules.

In conclusion, we have been able to demonstrate the confinement of 10^5 NH molecules in a magnetic trap with lifetimes exceeding 20 s. The long trap lifetimes are obtained by using a loading technique that creates a background gas density between 10^{11} and 10^{12} cm^{-3}. Trapping is accomplished in a cryogenic environment, avoiding constraints imposed by the optical pumping of molecules by blackbody radiation.

Acknowledgments

IOP Institute of Physics **Φ** DEUTSCHE PHYSIKALISCHE GESELLSCHAFT

References

[1] Hudson E R, Lewandowski H J, Sawyer B C and Ye J 2006 *Phys. Rev. Lett.* **96** 143004
[2] Bethlem H L, Kajita M, Sartakov B, Meijer G and Ubachs W 2008 *Eur. Phys. J. Spec. Top.* **163** 55
[3] DeMille D, Bay F, Bickman S, Kawall D, Krause D Jr, Maxwell S E and Hunter L R 2000 *Phys. Rev.* A **61** 052507
[4] Hudson J J, Sauer B E, Tarbutt M R and Hinds E A 2002 *Phys. Rev. Lett.* **89** 023003
[5] Lambert D L, Gustafsson B, Eriksson K and Hinkle K H 1986 *Astrophys. J. Suppl. Ser.* **62** 373
[6] Edvardsson D, Williams C F and Clary D C 2006 *Chem. Phys. Lett.* **431** 261
[7] Frankcombe T J and Nyman G 2007 *J. Phys. Chem.* A **111** 13163
[8] Stuhler J, Griesmaier A, Koch T, Fattori M, Pfau T, Giovanazzi S, Pedri P and Santos L 2005 *Phys. Rev. Lett.* **95** 150406
[9] DeMille D 2002 *Phys. Rev. Lett.* **88** 067901
[10] Andre A, DeMille D, Doyle J M, Lukin M D, Maxwell S E, Rabl P, Schoelkopf R J and Zoller P 2006 *Nat. Phys.* **2** 636
[11] Campbell W C, Tsikata E, Lu H-I, van Buuren L D and Doyle J M 2007 *Phys. Rev. Lett.* **98** 213001
[12] Campbell W C, Groenenboom G C, Lu H-I, Tsikata E and Doyle J M 2008 *Phys. Rev. Lett.* **100** 083003
[13] van de Meerakker S Y T, Vanhaecke N, van der Loo M P J, Groenenboom G C and Meijer G 2005 *Phys. Rev. Lett.* **95** 013003
[14] Lara M, Bohn J L, Potter D, Soldan P and Hutson J M 2006 *Phys. Rev. Lett.* **97** 183201
[15] Modugno G, Ferrari G, Roati G, Brecha R J, Simoni A and Inguscio M 2001 *Science* **294** 1320
[16] Avdeenkov A V, Kajita M and Bohn J L 2006 *Phys. Rev.* A **73** 022707
[17] Kajita M 2006 *Phys. Rev.* A **74** 032710
[18] Ketterle W and van Druten N J 1996 *Adv. At. Mol. Opt. Phys.* **37** 181
[19] Davis K B, Mewes M-O, Andrews M R, van Druten N J, Durfee D S, Kurn D M and Ketterle W 1995 *Phys. Rev. Lett.* **75** 3969
[20] Harris J G E, Michniak R A, Nguyen S V, Brahms N, Ketterle W and Doyle J M 2006 *Euro. Phys. Lett.* **67** 183201
[21] Brahms N 2008 *PhD Thesis* Harvard University
[22] Michniak R 2004 *PhD Thesis* Harvard University
[23] Herzberg G 1989 *Molecular Spectra and Molecular Structure* vol 1 (Malabar, FL: Krieger)
[24] Hoekstra S, Gilijamse J J, Sartakov B, Vanhaecke N, Scharfenberg L, van de Meerakker S Y T and Meijer G 2007 *Phys. Rev. Lett.* **98** 133001
[25] Sawyer B C, Stuhl B K, Wang D, Yeo M and Ye J 2008 *Phys. Rev. Lett.* **101** 203203
[26] Hummon M T, Campbell W C, Lu H-I, Tsikata E, Wang Y and Doyle J M 2008 *Phys. Rev.* A **78** 050702
[27] Patterson D and Doyle J M 2007 *J. Chem. Phys.* **126** 154307
[28] Maxwell S E, Brahms N, de Carvalho R, Glenn D R, Helton J S, Nguyen S V, Patterson D, Petricka J, DeMille D and Doyle J M 2005 *Phys. Rev. Lett.* **95** 173201
[29] Doret S C, Connolly C, Ketterle W and Doyle J M 2009 *Phys. Rev. Lett.* **103** 103005
[30] Stoll M, Bakker J M, Steimle T C, Meijer G and Peters A 2008 *Phys. Rev.* A **78** 032707