

for 1-GeV/nucleon  $^{16}\text{O}$  on Ag. In addition, if their cross section were spread uniformly over their observable energy range of 200 MeV/nucleon,  $d^2\sigma/d\Omega dE$  would be about 1 mb/MeV sr. Our curves for the Ag target (like those in Fig. 2 but down by a factor of  $2\frac{1}{2}$ ) are smooth in both energy and angle, and at  $40^\circ$  drop below this value at 35 and 60 MeV/nucleon for  $^4\text{He}$  and  $^3\text{He}$ , respectively. Thus we may conclude, without any assumptions about high-multiplicity stars, that the events of Baumgardt *et al.* are not dominantly due to high-energy He nuclei.

In conclusion, our data present evidence for the nonevaporative emission of  $^3\text{He}$  and, to a somewhat lesser extent,  $^4\text{He}$  products in collisions between relativistic heavy ions. The cross sections for these high-energy products are two to three orders of magnitude higher than those found for proton-induced reactions at comparable incident velocity.<sup>14</sup> This points towards a cooperative mechanism that cannot be explained by geometrical considerations or by an independent superposition of nucleon-induced knockon cascades.

We would like to thank Dr. W. Greiner, Dr. E. Schopper, Dr. J. R. Nix, and Dr. P. G. Siemens for useful discussions.

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<sup>14</sup>At incident proton energies of about 30 GeV, a high yield of  $^3\text{H}$  and  $^3\text{He}$  has also been observed that was interpreted as an emission from hadronic fireballs [R. Hagedorn and J. Ranft, Nuovo Cimento Suppl. **6**, 169 (1968)]. At our incident energies of about 1 GeV/nucleon, the center-of-mass energy of a *single* nucleon-nucleon scattering system is too low to allow for such emission. However, final-state interactions of cascade nucleons is a possibility.

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## Detection and Lifetime Measurement of Rb-He Quasibound Molecules\*

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Quasibound Rb-He Van der Waals molecules have been observed through their influence on the electron spin relaxation of optically pumped Rb<sup>85</sup> and Rb<sup>87</sup>. The natural lifetime of the complex has been measured to be  $6 \times 10^{-10}$  sec.

Atomic-beam and optical-pumping experiments have yielded considerable information on the properties of bound Van der Waals molecules formed in three-body collisions of alkali-metal atoms with noble-gas atoms.<sup>1-3</sup> Two-body or three-body collisions of alkali-metal with noble-gas atoms also should lead at times to the forma-

tion of molecular complexes which are quasibound, that is, to molecular states which have energies that are positive but less than the centrifugal-barrier potential.<sup>4-7</sup> In this Letter we report the first observation of a Rb-He quasibound molecular complex, together with an experimental determination of its mean lifetime

for dissociative tunneling through the centrifugal barrier and its cross section for collisional breakup. We find that much of electron spin relaxation of Rb previously attributed to sudden Rb-He collisions arises instead from the formation and destruction of Rb-He quasibound molecules.

The electronic spin relaxation induced in sudden binary collisions of alkali-metal atoms with noble-gas atoms can be represented in low magnetic fields by the sum of two exponential terms whose rate constants,  $Z_1$  and  $Z_2$ , differ by a factor dependent upon the magnitude of the nuclear spin.<sup>2</sup> The addition of wall relaxation, spin exchange, and pumping alters the magnitudes of these rate constants, but does not change the fundamental form of the optical-pumping transient:

$$\langle S_z \rangle_g = D_1 [1 - \exp(-Z_1 t)] + D_2 [1 - \exp(-Z_2 t)], \quad (1)$$

where  $\langle S_z \rangle_g$  is the electronic spin polarization of the alkali-metal vapor,  $D_1$  and  $D_2$  are functions of all pumping and relaxation rates, and weak pumping has been assumed.<sup>8</sup> Aymar, Bouchiat, and Brossel found that the measured ratios  $Z_2/Z_1$  for Rb in He were anomalously high compared to the theoretical predictions, but were unable to identify a cause of this effect.<sup>9</sup> Franz and Sooriamoorthi found that anomalous relaxation also exists for Cs in high pressures of noble gases, and argued that it was most likely due to the formation and destruction of Van der Waals molecules.<sup>8</sup> In the present paper we demonstrate that the anomalous relaxation which exists for Rb in He arises primarily from the formation of quasi-bound Rb-He molecules.

In Fig. 1 we present measurements of  $Z_2 - Z_1$  for Rb<sup>85</sup> in He, obtained in a white-light optical-pumping experiment. The experimental conditions were similar to those reported in Ref. 8 except that the cell temperature was electronically controlled at  $(305.0 \pm 0.1)^\circ\text{K}$  in the present case. The error bars represent  $\pm 1$  standard deviation of at least five measurements. A linear fit to the data yields a slope of  $0.0119 \text{ sec}^{-1} \text{ Torr}^{-1}$  and a zero-pressure intercept of  $13.9 \text{ sec}^{-1}$ . Theory predicts that  $Z_2 - Z_1$  should be dependent only upon  $\sigma$ , the nuclear-spin-independent cross section for the relaxation of  $\langle S_z \rangle_g$  in sudden alkali-atom-buffer-gas-atom collisions, and  $\sigma_s$ , the spin-exchange cross section:

$$Z_2 - Z_1 = \frac{17}{18} N_0 \sigma \bar{V}_{\text{rel}} p / p_0 + \frac{32}{54} N_0 \sigma_s \bar{V}_{\text{Rb-Rb}} p_{\text{Rb}} / p_0 + 0.6, \quad (2)$$

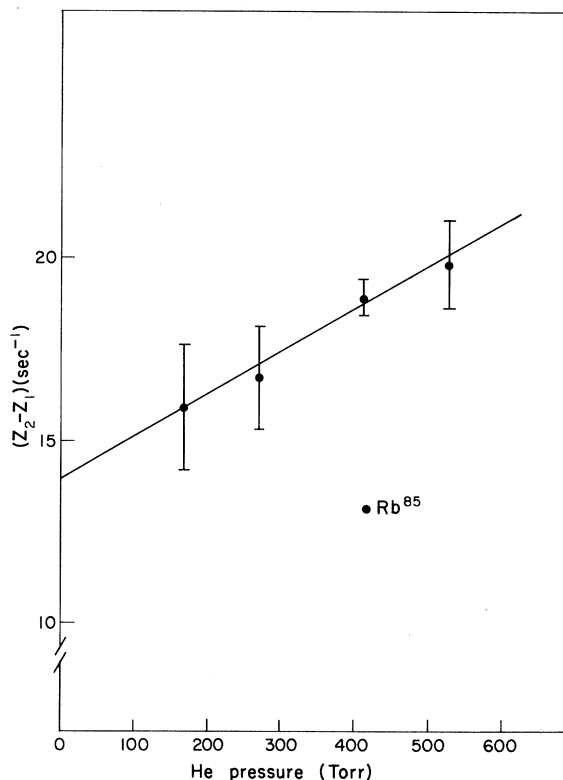


FIG. 1. Difference of the measured rate constants for weak optical pumping of  $\langle S_z \rangle$  of Rb<sup>85</sup> as a function of high He pressure.

where  $p$  is the buffer-gas pressure,  $p_{\text{Rb}}$  is the Rb<sup>85</sup> vapor pressure, and the rest of the parameters are defined in Ref. 8.<sup>10</sup> From the measured slope of  $Z_2 - Z_1$  versus  $p$  we obtain a value of  $(3.0 \pm 1.0) \times 10^{-24} \text{ cm}^2$  for  $\sigma$  for Rb-He. Taking  $\sigma_s$  to be  $2.0 \times 10^{-14} \text{ cm}^2$ ,<sup>11</sup> the saturated vapor pressure of Rb at  $305^\circ\text{K}$  to be  $3.1 \times 10^{-7} \text{ Torr}$ ,<sup>12</sup> and probable errors in each of these parameters to be less than 10%, we calculate that the extrapolated ( $p = 0$ ) intercept of  $Z_2 - Z_1$  should occur at approximately  $5.1 \pm 1.0 \text{ sec}^{-1}$ . In fact, however,  $13.9 \pm 2.0 \text{ sec}^{-1}$  was measured, about 3 times the anticipated result. Anomalous relaxation thus is present to a striking degree in the Rb-He system.

At low pressures of He  $Z_1$  and  $Z_2$  are predicted to be nearly equal, differing by an amount approximately equal to the spin exchange rate. When  $Z_2/Z_1$  is 1.5 or less the optical-pumping transient can be approximated by a single exponential with rate constant

$$Z_3 = (D_1 Z_1 + D_2 Z_2) (D_1 + D_2)^{-1}, \quad (3)$$

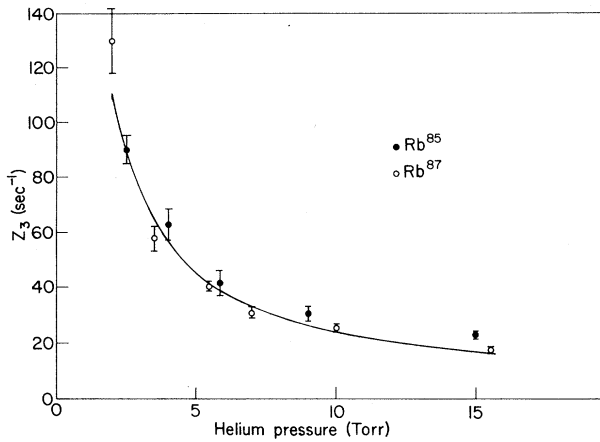


FIG. 2. Effective single exponential relaxation rates of  $\langle S_z \rangle_g$  of  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  at low He pressures.

where  $D_1$ ,  $D_2$ ,  $Z_1$ , and  $Z_2$  are the parameters of the true double-exponential transient.<sup>8</sup> In Fig. 2 we present data obtained from single-exponential fits of pumping transients of  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  in low pressures of He. The solid line is a best fit of Eq. (3) to the  $\text{Rb}^{87}$  data, yielding a value of 0.42 for  $D_0$ , the diffusion coefficient of Rb in He at 305°K. We note that the relaxation rates measured for  $\text{Rb}^{85}$  fall significantly above those measured for  $\text{Rb}^{87}$ . A small part of this difference is predicted from nuclear-spin considerations.<sup>8</sup> The major part of the excess observed, however, is anomalous. Bouchiat and co-workers studied somewhat similar effects in Rb-Kr, where bound molecules are formed, the correlation times for collisional relaxation at low Kr pressures are relatively long, and  $F$  remains a good quantum number throughout the relaxation events (only  $\Delta F = 0$  transitions occur). Theory and experiment show that for such a process larger relaxation rates are measured for  $\text{Rb}^{87}$  ( $I = \frac{3}{2}$ ) than for  $\text{Rb}^{85}$  ( $I = \frac{5}{2}$ ).<sup>2</sup> The experimental results for Rb-He displayed in Fig. 2, however, are just the opposite.

Although the individual data are not reproduced here, our measurements of  $Z_1$  and  $Z_2$  for  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  at He pressures ranging from 18 to 525 Torr indicate that throughout this pressure range anomalous relaxation makes a far smaller contribution to  $Z_1$  than to  $Z_2$ . The rate of anomalous relaxation appearing in  $Z_2$  must therefore depend mainly on transitions *between* hyperfine levels ( $\Delta F = \pm 1$ ) rather than on transitions *within* hyperfine levels ( $\Delta F = 0$ ).<sup>2,13</sup> We shall take the anoma-

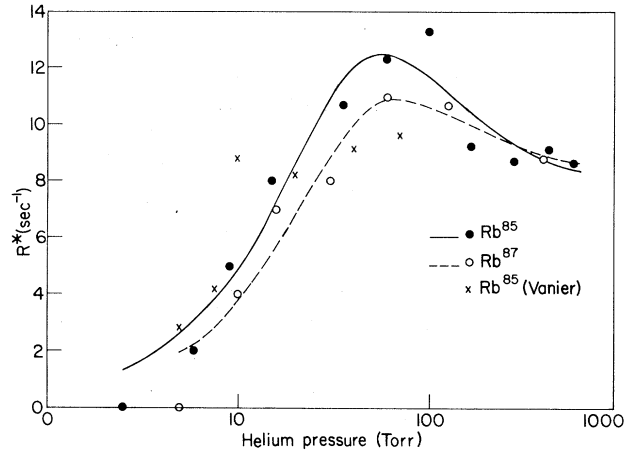


FIG. 3. Evaluated anomalous relaxation,  $R^*$ , in  $\text{Rb}^{85}$  and  $\text{Rb}^{87}$  as a function of He pressure with fits of Eq. (4) to the data. The  $\times$ 's represent our estimations of  $R^*$  from the experimental data of Vanier, Simard, and Boulanger (Ref. 15).

lous part of  $Z_2$  as essentially equal to the nuclear-spin-independent anomalous relaxation rate itself,  $R^*$ . The results of such reductions of data at He pressures from 18 to 525 Torr are displayed in Fig. 3. With somewhat more effort it is possible to find the value of  $R^*$  necessary to make the calculated values of  $Z_3$  equal to the single-exponential fits to low-pressure data. Such determinations provide the data points from 2.5 to 13 Torr in Fig. 3. The estimated uncertainties on all points are approximately 2  $\text{sec}^{-1}$ .

The fact that  $R^*$  becomes constant at high He pressures indicates that anomalous relaxation cannot be attributed to effects arising from normal fly-by binary collisions: Relaxation rates arising from such a source would be proportional to He pressure. The observed ultimate fall of  $R^*$  at low He pressure substantiates the earlier demonstration that anomalous relaxation is not related to spin exchange: Spin-exchange effects would be independent of He pressure.<sup>8</sup> We now shall show that all of the observed characteristics of  $R^*$  can be explained by considering the contributions to relaxation rates from the formation of *quasibound* Van der Waals molecules.

Slichter<sup>14</sup> has calculated the relaxation rate for a spin- $\frac{1}{2}$  particle subjected to a magnetic field which jumps randomly between two fixed values. A simple adaptation of Slichter's calculation to Rb spin relaxation induced by quasimolecular formation leads to the following equa-

tion<sup>13</sup> for  $R^*$ :

$$R^* = A\tau_c^2(1 + \omega_0^2\tau_c^2)^{-1}, \quad (4)$$

where

$$\tau_c^{-1} \simeq \tau^{-1} + N_0\sigma_B V_{rel} p/p_0. \quad (5)$$

$A$  is a factor directly proportional to the rate of formation of complexes,  $\omega_0$  is the resonant frequency for the electron spin polarization (in our case  $2\pi$  times the hyperfine frequency),  $\tau$  is the natural lifetime of the quasibound complex, and  $\sigma_B$  is the cross section for collisional breakup. Bouchiat, Bouchiat, and Pottier also derived Eq. (4) in their work on Rb-heavy-noble-gas Van der Waals molecules; in that case the formation rate was proportional to  $p^2$  since three-body collisions are required for the formation of bound states.<sup>2</sup> In our case we expect  $A$  to be of the form

$$A = Bp + Cp^2, \quad (6)$$

where  $Bp$  is proportional to the formation rate of quasibound complexes in two-body collisions, and  $Cp^2$  is proportional to the formation rate of quasibound plus bound complexes in three-body collisions. The case of Rb-He should be particularly interesting in that we calculate that there should be only one bound (twofold degenerate) and one quasibound (sixfold degenerate) vibrational state<sup>4,7</sup>: Even three-body collisions will lead mostly to the formation of quasibound molecules.

The solid line in Fig. 3 is a fit of Eqs. (4)–(6) to the  $R^*$  data for Rb<sup>85</sup>; the dashed line is a fit to the data for Rb<sup>87</sup>. For Rb<sup>85</sup> we obtain  $\tau = 6 \times 10^{-10}$  sec and  $\sigma_B = 1 \times 10^{-13}$  cm<sup>2</sup>; for Rb<sup>87</sup> we obtain  $\tau = 3 \times 10^{-10}$  sec and  $\sigma_B = 3 \times 10^{-13}$  cm<sup>2</sup>. We believe our Rb<sup>85</sup> results to be the more reliable, but because of the roughness of the data we must consider them accurate only to within a factor of 2 or so. Our value of  $\tau$  is the same order of magnitude as that calculated by Baylis for the highest quasibound states of Rb-Ar.<sup>5</sup> The ratio  $B/C$  was 95 Torr for Rb<sup>85</sup> and 63 Torr for Rb<sup>87</sup>. Qualitatively we see that at low pressures the term  $Bp$  dominates, indicating that complexes are being formed primarily in two-body collisions. At high pressures the term  $Cp^2$  dominates, indicating that three-body collisions are dominant in complex formation in that regime, forming either bound or quasibound complexes. At a pressure of about 100 Torr the complex formation rates in two- and three-body collisions are approximately equal. Referring to Eq. (4) we see that the difference in relaxation rates of Rb<sup>85</sup> and Rb<sup>87</sup> at

low pressures arises primarily from the fact that  $\omega_0(\text{Rb}^{85}) < \omega_0(\text{Rb}^{87})$ .

It is now seen that far more of the relaxation rate of  $\langle S_z \rangle$  of Rb in He arises from the formation of Rb-He quasibound complexes than from sudden Rb-He binary encounters. Analysis of experiments made without this knowledge leads to erroneously high values for  $\sigma$ , such as the value of  $8.2 \times 10^{-24}$  cm<sup>2</sup> for  $\sigma(\text{Rb-He})$  reported by Aymar, Bouchiat, and Brossel. A similar effect exists in the recent report of Vanier, Simard, and Boulanger.<sup>15</sup> These authors used their Rb<sup>85</sup> maser to measure relaxation rates of  $\langle \vec{S} \cdot \vec{I} \rangle$  for Rb<sup>85</sup> in various noble gases at pressures up to 70 Torr. In fitting theoretical curves to experimental data they allowed the spin exchange rate,  $R_s$ , to be a free parameter along with  $D_0$  and  $\sigma$ . While they do not quote specific values of  $R_s$  thus obtained, inspection of their published curves indicate that a value of approximately  $15.6 \text{ sec}^{-1}$  is required to fit their Rb-He curve. However, we estimate that  $R_s$  should have been at most  $4.5 \text{ sec}^{-1}$  under the conditions of their experiment. This large difference is caused by the misattribution of the relaxation rate from the formation of quasibound complexes to spin exchange. Reanalysis of the data of Vanier, Simard, and Boulanger in the light of present knowledge yields values of anomalous relaxation rates which lie very close to the solid curve in Fig. 3, in good accord with the new theoretical expectation. Finally, we note that the value of  $3.0 \times 10^{-24}$  cm<sup>2</sup> for  $\sigma$  reported here is in reasonable agreement with the theoretical calculations of  $1.9 \times 10^{-24}$  cm<sup>2</sup> made by Dashevskaya and Kobzeba<sup>16</sup> and  $3.8 \times 10^{-24}$  cm<sup>2</sup> made by Herman.<sup>17</sup>

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## Localized Fields and Density Perturbations Due to Self-Focusing of Nonlinear Lower-Hybrid Waves\*

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Large-amplitude lower-hybrid wave bursts at  $\omega_0 > \omega_{LH}$  are excited from a grid and propagate into a density gradient perpendicular to  $\vec{B}_0$ . The evolution of the internal rf fields and the plasma parameters is investigated. Above threshold localized field maxima appear ( $E_{int} \gg E_{app}$ ) and, simultaneously, strong density perturbations are formed. The nonlinear phenomena are interpreted by ponderomotive-force and space-charge separation effects.

Nonlinear wave phenomena are a topic of strong current interest. The ponderomotive force in strong nonuniform rf fields can change the local plasma parameters which may enhance the field and lead to instabilities. Filamentation instabilities of plane waves have been discussed in the literature for both unmagnetized<sup>1</sup> and magnetized plasmas.<sup>2</sup> The present work is concerned with a filamentation process of an initially nonuniform rf field, i.e., the resonance cone pattern<sup>3</sup> associated with lower-hybrid waves. Locally growing density depressions and internal rf fields are observed.

The experiment is performed in a large (12 cm diam, 150 cm length), quiescent ( $\delta n/n \approx 1\%$ ), weakly collisional ( $\nu_{en}/\omega < 10^{-2}$ ), magnetized ( $B_0 \approx 500$  G) plasma column ( $n_e \approx 10^{10}$  cm<sup>-3</sup>,  $T_e \approx 2$  eV) described in detail elsewhere.<sup>4</sup> In a previous experiment<sup>5</sup> we studied the three-dimensional propagation of small-amplitude lower-hybrid waves in the uniform center region of the column ( $\sim 8$  cm diam). By suitably modifying the cathode, we have now established a constant density gradient ( $n/\nabla n \approx 15$  cm over 8 cm across  $\vec{B}_0$ , see Fig.

1) and are able to launch waves from the low-density side with  $\vec{k} \perp \nabla n$ ,  $\vec{k} \perp \vec{B}_0$ . The exciter grid is 66 cm long, 3.5 cm high, and consists of fourteen electrically connected, plane parallel to  $\vec{B}_0$ , spring-loaded, 0.1-mm-diam tungsten wires. With this design we minimize density perturbations yet establish an equipotential surface in the plasma, which closely matches the phase front of lower-hybrid normal modes. The waves are detected with  $T$  probes (3 cm length, 0.1 mm diam, 1-mm-o.d. coaxial feed), movable axially along  $\vec{B}_0$  and in the radial directions (parallel and perpendicular to the exciter-grid surface normal, respectively). Phase-coherent wave bursts ( $4 < f < 20$  MHz,  $0.05 < t < 10$   $\mu$ sec,  $1 < V_0 < 60$  V peak to peak at  $Z = 50$   $\Omega$ ) are applied to the exciter grid. The received signals are analyzed with a sampling oscilloscope and/or boxcar integrator.

The propagation of lower-hybrid waves along a density gradient reveals a more complicated structure than in a uniform plasma. Figure 1 shows backward lower-hybrid waves near the exciter bounded by a standing wave corresponding