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Absence of Quasibound Negative-Ion Ground States of He and H_2 in Electron Scattering*

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A search for quasibound negative-ion ground states in electron scattering from He and H_2 in transmission has been made with negative results. The half-width of the electron beam used in these experiments was about 0.05 eV and was independent of electron energy in the interaction region. The lower limit for detection of cross-section changes in this experiment was about 2 or 3 parts in 10^4. The series of elastic vibrational resonances in N_2 which are probably due to the quasibound negative-ion ground state of N_2 are discussed.

INTRODUCTION

Since the finding of the elastic core-excited resonance in e^--He scattering at about 19.3 eV,[1] many core-excited resonances have been found in both the elastic and inelastic channels of a variety of atoms and molecules by electron scattering.[2–9] All of these resonances have been found in the old channel approximately 0.5 eV below the threshold of the new channel. For example, the 19.3-eV helium resonance can be thought of as being due to a short-lived state of He^- with an electronic configuration of $1s$, $2s^2$. If, however, this electronic configuration is attributed to the 19.3-eV helium resonance, then the possibility of a state of He^- with the electronic configuration of $1s^2$, $2s$ and energy somewhere between 0 and 19.3 eV should be considered.

Since a bound state of H^- ($1s^2$) is known to exist, the simplest system in which to look for a non-core-excited resonance is e^--He. The system e^--H_2 must also be considered since the molecular system would have a more complicated excitation mechanism.

"Fine structure" has been observed in the early measurements of low-energy e^--He cross sections by both Ramsauer and Kollath[10] and Normand.[11] The more recent experiment of Golden and Bandel[12] did not disclose any "fine structure" in e^--He scattering. Recently, Schulz[13] has reported a strong resonance structure in e^--He scattering centered at 0.45 eV (similar to that observed by Ramsauer and Kollath[10] and Normand,[11] but not as broad) by observing the scattered electron current at 72°.

The data of Golden and Bandel[12] in He were obtained as cross-section values at various values of incident electron energy. Therefore, the possibility exists that the experiment of Golden and Bandel[12] could have missed a very sharp resonance. Although a modification of the Ramsauer technique[9] allowed a continuous variation of electron energy for energies greater than 1.5 eV, this technique is not suitable for lower electron energies. Hence, it was decided to build a new apparatus with high sensitivity for cross-section changes, good energy resolution, and the capability of continuous energy variation for low electron energies in order to resolve the question of the existence or nonexistence of quasibound negative-ion ground states of He and H_2 in electron scattering.

APPARATUS

A schematic diagram of the apparatus used in this work is shown in Fig. 1. The chamber is of all-metal construction and is connected through a molecular-sieve trap[14] to an "expanded" 4-in. oil diffusion pump which contains an "Edwards cold cap." The apparatus is part of an ultra-high-vacuum system which is capable of an ultimate pressure of less than 10^{-9} Torr.[15,16] All of the metal parts of the chamber which are visible to the electron beam have been sprayed with a dispersion of colloidal graphite in order to decrease contact potential differences between various parts.[17] All insulators have been placed so they are not visible to the beam. The electrostatic selector is of the 127° cylindrical type and is similar to that described by Hughes and McMillen.[18] Electrons from an indirectly heated cathode are accelerated by a negative voltage V_k on it with respect to the apparatus which is grounded. The grid is operated either positive or negative with respect to

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[1] G. J. Schulz, Phys. Rev. Letters **10**, 104 (1963).
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[9] D. E. Golden and H. W. Bandel, Phys. Rev. Letters **14**, 1010 (1965).
[10] C. Ramsauer and R. Kollath, Ann. Physik **3**, 536 (1929).
[11] C. E. Normand, Phys. Rev. **35**, 1217 (1930).
[12] D. E. Golden and H. W. Bandel, Phys. Rev. **138**, A14 (1965).
[13] G. J. Schulz, *Abstracts of the Fourth International Conference on the Physics of Electronic and Atomic Collisions, Quebec, 1965* (Science Bookcrafters, Hastings-on-Hudson, New York, 1965).
[14] M. A. Biondi, Rev. Sci. Instr. **30**, 831 (1959).
[15] During bakeout the gas inlet tube is connected to the pump-out tube in order to facilitate evacuation of the chamber behind the differential-pumping slit.
[16] With differential pumping, the ratio of pressure in the interaction region to that in the gun region is about 40.
[17] J. H. Parker and R. W. Warren, Rev. Sci. Instr. **33**, 948 (1962).
[18] A. L. Hughes and J. H. McMillen, Phys. Rev. **34**, 291 (1929).

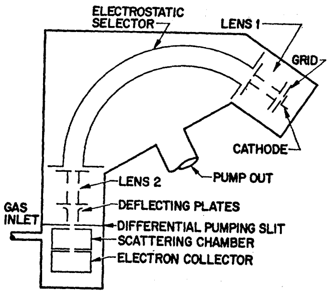

FIG. 1. The experimental arrangement.

the cathode, as needed to obtain suitable currents and beam conditions. Suitable voltages are applied to the electrodes of lens 1 to focus a beam of electrons into the entrance of the electrostatic selector. The electrostatic selector is operated with a positive voltage V_1 on the inner electrode and a negative voltage V_2 on the outer electrode with respect to ground such that $V_1 \gtrsim V_2$.[19] Suitable voltages are applied to the electrodes of lens 2 to focus the beam emerging from the electrostatic selector, through the scattering chamber and into the electron collector. A small voltage difference may be applied between the deflecting plates in order to correct minor beam-alignment difficulties. The dimensions will be given in inches. The grid slit and the slits in lenses 1 and 2 are 0.039×0.394. The grid is 0.125 from the first slit of lens 1. Lenses 1 and 2 are 0.3×1.25 and are 0.25 from the entrance and exit of the electrostatic selector, respectively. The inner and outer radii of the selector are 1.969 and 2.362, respectively. The deflecting plates are 0.75 long and 1.0 high and are 0.05 from both lens 2 and the differential-pumping slit. The differential-pumping slit is 0.050×0.500. The scattering chamber is 1.00 long and is 0.050 from the differential-pumping slit. The scattering-chamber and electron-collector slits are 0.060×0.500. The electron collector is 0.150 from the scattering chamber and has a ratio of slit area to inside surface area of about 0.007. The pressure in the interaction region is measured by a Schulz-Phelps high-pressure ion gauge.[20] The gas-handling system allows for the introduction of two gases at one time into the interaction region.

The energy of the electron beam is given by

$$E \simeq 2.75(V_1 - V_2), \tag{1}$$

where V_1, V_2, and E are in volts. However, the voltage drop across the oxide-coated cathode is such that

$$V_k(\text{volts}) \simeq E + 2. \tag{2}$$

PROCEDURE

The gun is operated so as to allow a beam of low-energy electrons (usually between about 0.7 and 1.3 eV) to reach the electron collector. The energy of the electron beam is determined by making retarding potential measurements on the collector. Figure 2 shows a plot of a portion of a collector current I_c versus retarding potential V_c in vacuum, and the energy distribution obtained therefrom by graphical differentiation. The half-width obtained from this plot is about 0.05 eV. In general in the energy range used in this work, the half width of the beam was always 0.05 ± 0.005 eV.[21,22]

The energy spread of the beam ΔE as determined by the slit system is roughly given by

$$\Delta E/E \simeq \Delta r/r, \tag{3}$$

where Δr is the slit width of the selector slits and r is the mean radius of the selector. However, for sufficiently low values of electron energy the energy spread of the beam reaches a limiting value which in this apparatus is about 0.045 eV.

In order to vary the electron energy, a suitable potential was applied to the scattering chamber and the collected current was measured as a function of this voltage while keeping the pressure in the interaction region constant. The energy in the interaction region was determined by adding (or subtracting) the potential applied to the scattering chamber to the initial energy as determined from the retarding-potential measurements.

The collected current $I_c(E)$ may be written

$$I_c(E) = I_{c0}(E) \exp[-\sigma_t(E) n x], \tag{4}$$

where $I_{c0}(E)$ is the collected current at zero pressure, $\sigma_t(E)$ is the total scattering cross section, n is the gas

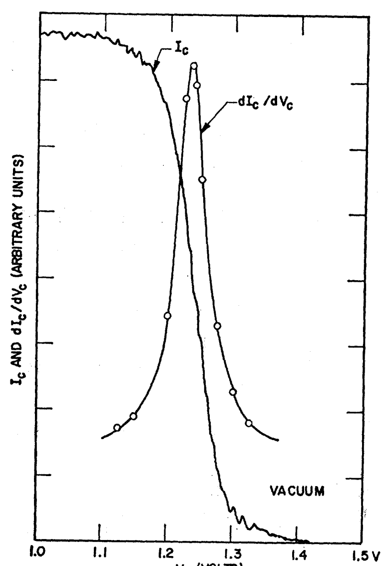

FIG. 2. A typical retarding-potential plot and electron energy distribution obtained therefrom by graphical differentiation.

[19] M. G. Inghram and R. J. Hayden, Natl. Acad. Sci.—Natl. Res. Council, Nucl. Sci. Ser. No. 14 (1954).

[20] G. J. Schulz and A. V. Phelps, Rev. Sci. Instr. **28**, 1051 (1957).

[21] Retarding-potential measurements on the electron beam in the presence of gas at operating pressures yielded the same shape and energy spread as measurements in vacuum.

[22] When the electron beam was centered in the slit system, retarding-potential measurements at the collector or at the scattering chamber were essentially the same.

density, and x is the path length of the electron beam through the scattering chamber. If $I_{c0}(E)$ is assumed to be a very slowly varying function of E as compared to the variation in $I_c(E)$ due to a resonance, the change in current in the vicinity of a resonance $\delta I_c(E)$ may be written

$$\delta I_c(E)/I_c \simeq [\delta\sigma(E)/\sigma_t](\sigma_t nx), \tag{5}$$

where $\delta\sigma$ is the change in cross section in the vicinity of a resonance which is assumed to be large compared to the variation of $\sigma_t(E)$ in the same vicinity. Equation (5) allows an approximate determination of the percentage change in cross section at a resonance providing one has knowledge of the total cross section at that energy.

Equation (5) is not valid for multiple scattering, and hence the pressure in the interaction region was always adjusted to keep $\sigma_t nx \leqslant 3$ in order to minimize the multiple-scattering problem.

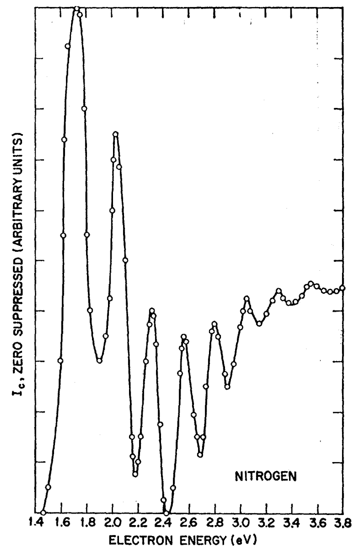

FIG. 4. Points obtained from a series of recorder plots of I_c versus electron energy from about 1.4 to 3.8 eV in nitrogen run at a pressure of 0.010 Torr. The position of the peaks in I_c are reproducible to about ± 0.03 eV and the absolute energy scale is probably good to ± 0.1 eV.

RESULTS

Using the technique described above, the strong resonance in He was observed many times with the peak in the transmitted current at 19.27 ± 0.08 eV and with a half-width of 0.05 ± 0.005 eV. A recorder tracing of this resonance is shown in Fig. 3. The half-width on this particular plot is about 0.045 eV, and the maximum in the transmitted current I_c represents a cross-section change of about 18% using the data of Golden and Bandel for σ_t.[12] Helium was always one of the two gases attached to the gas handling system, and the position and width of this resonance was determined during each run for the other gases used. Comparisons between the width of this resonance and the beam width as determined from retarding-potential measurements were always in excellent agreement provided that the beam was centered in the slit system.

In order to test the ability of the apparatus to detect resonances for low values of electron energy, it was decided to look for the series of elastic resonances in N_2 starting at about 1.8 eV.[23] The results of these measurements are shown in Fig. 4. Eight broad resonances are observed, the positions and spacings of which are shown in Table I, together with the vibrational-spacings of the ground state of $N_2(^1\Sigma_g{}^+)$.[24] This state has been designated $^2\Pi_g$ by Gilmore.[25] It should be noted that the envelope of these resonances can be made to look quite different from that shown in Fig. 4 depending on the voltages on the lenses. In fact the lenses may be adjusted so as to eliminate one or more of the higher energy resonances. Within the experimental error involved, the spacings of these resonances are in good agreement with the spacings of the $^1\Sigma_g{}^+$ state and are

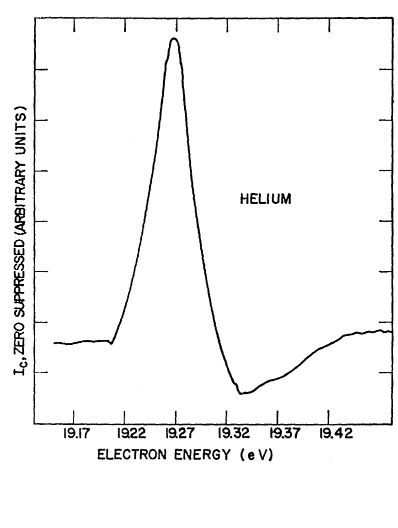

FIG. 3. A recorder tracing of I_c versus electron energy in the vicinity of the strong helium resonance at 19.27 eV run at a pressure of 0.040 Torr.

TABLE I. Observed position of resonances in N_2, and a comparison between the observed resonance spacings and the vibrational spacings of the ground state of N_2.

Observed transmission peak (eV)	Spacing (eV)	Vibrational spacing $^1\Sigma_g{}^+$
1.73		
	0.30	0.29
2.03		
	0.28	0.29
2.31		
	0.25	0.28
2.56		
	0.24	0.28
2.80		
	0.25	0.28
3.05		
	0.25	0.27
3.30		
	0.25	0.27
3.55		

[23] Four of these resonances have previously been observed by G. J. Schulz, Phys. Rev. 135, A788 (1964).

[24] The vibrational spacings for the $^1\Sigma_g{}^+$ were obtained from G. Herzberg, *Molecular Spectra and Molecular Structure* (D. Van Nostrand Company, Inc., New York, 1950) 2nd ed., Vol. 1.

[25] F. R. Gilmore, The Rand Corporation, Memorandum RM-4034-PR, 1964 (unpublished).

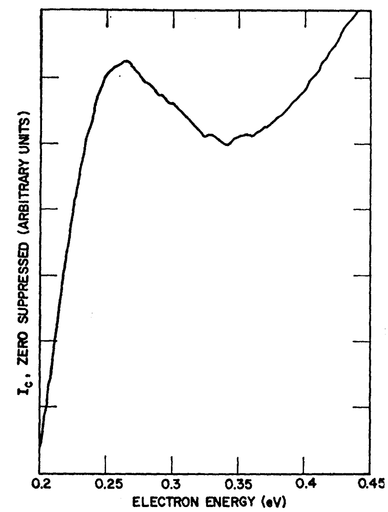

FIG. 5. A recorder tracing of I_c versus electron energy from 0.2 to 0.45 eV in nitrogen run at a pressure of 0.02 Torr. This plot shows a false "resonance" at about 0.26 eV.

probably an example of a quasibound negative-ion ground state.[26] It is rather difficult to apply Eq. (5) in the case of N_2 in this energy region due to the fact that σ_t is rapidly varying.[11] Furthermore, the well-known electron-optics effects introduce further complications. However, Eq. (5) may be used to give an estimate for the strength of these resonances. Such an estimate yields a value of 2×10^{-17} Å^2 for each division on the figure.

Using the same technique as described above, a systematic search was made between about 0.1 and about 19.3 eV in He and between about 0.1 and about 11.0 eV in H_2 for structure in the respective cross sections.[27] No structure was observed. The fact that a quasibound negative-ion ground state of H_2 was not found might have been expected. The ground state of $H_2^-(^2\Sigma_u^+)$ has been calculated to be repulsive.[28] This has been discussed by Chen and Magee.[29]

In order to test the necessity of alignment of the beam emerging from lens 2 in the center of the remaining slit system, a few hundredths of a volt additional potential difference was applied between the deflecting plates after the beam had been aligned in order to decrease the current reaching the collector. A plot of I_c versus electron energy in the interaction region as a result of this misalignment is shown in Fig. 5. A peak appears which can be shown to be false. This measurement was actually made with N_2 in the interaction region so that this "resonance" is about 0.05 eV wide and represents a change in cross section of about three parts in 10^3. While this effect was found with gas in the interaction region, it was found possible to obtain such an effect in vacuum. Nevertheless, when care was taken to peak the beam current into the collector with a small voltage between the deflecting plates, no such effect was observed.

The sensitivity for detection of cross-section changes may be determined from Eq. (5). The signal-to-noise ratio of the apparatus is such that a change in current of about 0.3% has a signal-to-noise ratio of about 10; using an attenuation factor $(\sigma_t nx)=3$ gives a conservative estimate of the smallest detectible change in cross section of $\delta\sigma/\sigma_t \simeq 2\times10^{-4}$.

[26] The observed spacings would also fairly well fit the vibrational spacings of the ground state of $N_2^+(^2\Sigma_g^+)$, indicating that the binding of the two N atoms is in large part due to only one valence electron.

[27] Values of σ_t for H_2 were obtained from D. E. Golden, H. W. Bandel, and J. A. Salerno (to be published).

[28] I. Fischer-Hjalmars, Arkiv Fysik **16**, 33 (1959).

[29] J. C. Y. Chen and J. L. Magee, J. Chem. Phys. **36**, 1407 (1962).