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# Investigation of excitation transfer in the system He($2^3S_1$) +Ne($2^1S_0$) by the method of optical orientation of atoms

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The method of optical orientation of atoms has been used to investigate the process of excitation transfer from helium atoms in the triplet metastable state to neon atoms. It turns out that this process has a cross section of $\sigma = 0.35$ Å$^2$ at $T = 300$ K and $\sigma = 0.06$ Å$^2$ at $T = 178$ K. Analysis of the experimental data within the framework of the Rosen-Zener-Demkov model allows the determination of the He–Ne system's exchange–interaction potential, which has the form $\Delta = 8000 \exp(-1.38R)$.

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## 1. INTRODUCTION

Metastable triplet helium atoms with an excitation energy of 19.82 eV can ionize by collisions at thermal relative-motion energies all atoms except neon, whose ionization potential is 21.56 eV. But the metastable helium atoms lose their excitation in the neon case too. This happens as a result of the transfer of the excitation energy of the $2^3S_1$ helium state to the nearby $2s_2$ and $2s_3$ levels of neon (electronic configuration $2p^54s$):

$$\mathrm{He}(2^3S_1)+\mathrm{Ne}(2^1S_0)\rightarrow\mathrm{Ne}(2s_2,\ 2s_3)+\mathrm{He}(1^1S_0). \tag{1}$$

This process is one of the principal processes that occur in the course of pumping in an infrared helium–neon laser, and quite a large number of papers have been devoted to the determination of its reaction rate.[1-10] The study of this reaction is also of interest from the point of view of the fact that it is not a strictly resonance process: the $2s_2$ and $2s_3$ neon levels respectively lie 0.039 and 0.058 eV below the $2^3S_1$ helium level.

Apropos of the data available in the literature on the magnitude of the reaction rate of the excitation-transfer process (1), we should note two facts:

1) The experiments were performed largely at room temperature. A summary of the data obtained by the various authors is given in Table I, where we give the values of the reaction rate and cross section, given, as usual, by the formulas

$$\sigma=C/2^{1/2}\bar{v},\ \bar{v}=(8kT/\pi\mu)^{1/2}, \tag{2}$$

where $\mu$ is the reduced mass of the He–Ne system. It can be seen from Table I that the data of the various authors differ considerably from each other.

Temperature investigations have been carried out by Jones *et al.*[5] in the 77-400-K temperature range, Lindinger *et al.*[7] in the 300-800 K range, and Arathoon[8] in the 140-340 K range. It turns out that the reaction rate decreases sharply with decreasing temperature. At the same time, these authors report significantly differing values for both the reaction rate and its rate of change with temperature.

2. In all the experiments, except Beterov and Chebotaev's,[4] the afterglow of a decaying helium – neon plasma was studied. The reaction rate of the process (1) was found by comparing the decay constants of the $2^3S_1$ helium state (i.e., the rates of decrease of the metastable triplet helium atom concentration in the afterglow) for pure He and a He–Ne mixture. The concentration of the He*($2^3S_1$) atoms in the afterglow is determined by

TABLE I. Results of experimental investigations of the process of excitation transfer He* to Ne at $T$ = 300 K.

| | $C$, $10^{-11}$ cm$^3$-sec$^{-1}$ | $\sigma$, Å$^2$ |
|---|---|---|
| Phelps[1] | 0.31 | 0.22 |
| Javan *et al.*[2] | 0.51±0.07 | 0.37±0.05 |
| Benton *et al.*[3] | 0.39±0.08 | 0.28±0.06 |
| Beterov and Chebotaev[4] | 0.52 | 0.38 |
| Jones *et al.*[5] | 0.43±20% | 0.31±20% |
| Schmeltekopf *et al.*[6] | 0.385±30% | 0.28±30% |
| Lindinger *et al.*[7] | 0.36±30% | 0.26±30% |
| Arathoon[8] | 0.72 | 0.52 |
| Lee and Collins[9] | <0.7 | <0.51 |
| Ernie and Oskam[10] | 0.43 | 0.31 |

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several processes involving the production and destruction of these atoms.[11] In Refs. 1-3, 5-10 it is tacitly assumed that these processes do not depend on the presence of Ne, and that the addition of it lead only to a new decay process for the He* atoms. But it is not clear how well this assumption is fulfilled, since the addition of neon also affects the processes leading to the production of He* in the decaying plasma.

Beterov and Chebotaev[4] used for the determination of the reaction rate of the process (1) an original procedure based on an analysis of the variation of the power of the helium–neon laser with decreasing concentration of the He* atoms: the decrease led to the decrease of the population of the $2s_2$ and $2s_3$ neon levels. But the population of these levels could have varied not only as a result of the direct process (1), but also as a result of various secondary processes occuring in the helium–neon plasma. Thus, it is clear that it would have been interesting to carry out the investigation of the reaction (1) in a fairly broad range of temperatures with the aid of a method significantly different from the methods used earlier.

In the present paper we employ the method of optical orientation of atoms, which allows us to carry out a practically direct measurement of the reaction rate of the process of excitation transfer from helium to neon. The information obtained is used to analyze qualitatively the interaction in the He*–Ne system within the framework of the well-known—in the thoery of atomic collisions—Rosen–Zener–Demkov model.[12]

## 2. EXPERIMENTAL DETERMINATION OF THE REACTION RATE FOR THE PROCESS OF EXCITATION TRANSFER FROM He* TO Ne

The optical orientation of helium atoms consists in the transfer of angular momentum from circularly polarized light to an ensemble of helium atoms in the metastable triplet state.[13] The optical orientation of the magnetic moments of the (spin $S = 1$) He* atoms was effected by the resonance radiation of a helium pumping lamp ($\lambda = 10830$ Å; the $2^3S_1 \rightarrow 2^3P$ transition). The experimental setup is described in detail in Ref. 14.

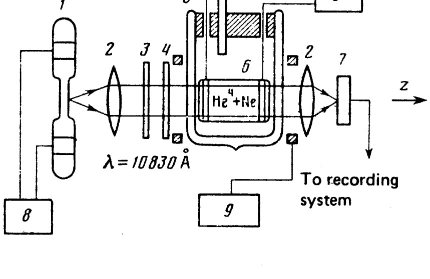

FIG. 1. Block diagram of the experimental setup for the optical orientation of the helium atoms in a $He^4$ + Ne mixture. 1) Capillary pumping helium lamp; 2) lenses; 3) PPI-1 polaroid; 4) quarter-wave mica plate; 5) Dewar vessel; 6) absorption cell; 7) FD-7K silicon photodetector; 8) high-frequency generator; 9) radio-frequency magnetic-resonance generator.

The absorption cell, filled with either $He^4$ (0.36 mm Hg at 300 K), or a mixture of $He^4$ (0.36 mm Hg) and Ne (from 0.053 to 0.252 mm Hg), was placed in a uniform constant magnetic field, $H_0 \sim$ Oe, directed along the pumping beam (Fig. 1). A sufficient concentration ($\sim 10^{10}$ cm$^{-3}$) of the metastable He* atoms was attained as a result of the use of a high-frequency gas discharge in the absorption cell. A magnetic resonance was realized in the system of Zeeman sublevels of the $2^3S_1$ state of helium with the aid of an oscillating magnetic field directed perpendicularly to $\mathbf{H}_0$. The resonance signals were recorded from the absorption of 1.08-μm pumping light.

In the experiments we determined the magnetic-resonance line width of the metastable triplet helium atoms in the temperature range from 178 to 300 K with a temperature-point stability of better than 3 K. The temperature was measured on the surface of the absorption cell. This temperature differed only insignificantly (within the limits from 1 to 3 K) from the mean temperature inside the absorption cell. This is due to the fact that we used a very weak high-frequency discharge. The intensity measurements performed on this discharge did not lead to a change in the line width, which change was to be expected in the case of appreciable heating of the plasma of the helium–neon discharge, considering the strong temperature dependence of the reaction rate of the process (1). Furthermore, Colegrove *et al.*[15] have shown that even under poor heat-transfer conditions the mean temperature of the absorption cell does not exceed the surface temperature by more than 3 K in optical-helium-atom-orientation experiments in the temperature range from 4.2 to 500 K.

We determined the reaction rate of the excitation-transfer process (1) on the basis of an analysis of the resonance line width of the He* atoms. This analysis embraces three stages:

1) The determination of the magnetic-resonance line width, $\Delta f_{\mathrm{He+Ne}}$, of the He* atoms in a $He^4$ + Ne mixture upon the extrapolation to zero of the discharge and pumping-light intensities, the amplitude of the radio-frequency magnetic field, and the modulation amplitude of the constant magnetic field;

2) the determination of the quantity $\Delta f_{\mathrm{He}}$ under the same experimental conditions, but in the presence in the absorption cell of only the pure isotope $He^4$;

3) the separation of the contribution, due to the addition of neon, to the resonance line width, and the determination of the reaction rate of the excitation-transfer process (1) on the basis of the relation

$$\pi\Delta f_{\mathrm{Ne}}=N_{\mathrm{Ne}}C, \tag{3}$$

where $N_{\mathrm{Ne}}$ is the neon concentration, $C = \langle \sigma v \rangle$ is the reaction rate of the process (1), $v$ is the relative He*–Ne collision rate, and $\Delta f_{\mathrm{Ne}}$ is the contribution made by the reaction (1) to the resonance line width, and given by the relation

$$\Delta f_{\mathrm{Ne}}=\Delta f_{\mathrm{He+Ne}}-\Delta f_{\mathrm{He}}+\delta f_D, \tag{4}$$

where $\delta f_D$ is the diffusional correction term, which will be discussed in greater detail below.

To attain a higher accuracy in the determination of the magnitude of the reaction rate, it is desirable to have several absorption cells with different neon concentrations, including the highest possible concentrations. But the increase of the quantity of neon in the absorption cell is limited by the fact that it leads, first, to the decrease of the He* concentration and, second, to the increase of the rate of depolarization of the He atoms in the $2^3P$ state, as a result of which the magnetic resonance signals decrease sharply. The minimum Ne concentration is determined by the smallest observable contribution to the magnetic resonance line width of He*. Therefore, we were able to use at each temperature a set of (up to 5) absorption cells in which the Ne content varied from 0.053 to 0.252 mm Hg (when calculated at 300 K).

The quantity $\Delta f_{\mathrm{Ne}}$, as defined by the relation (4), can be determined in much the same way as the broadening of the resonance line of helium in the system $\mathrm{He}(2^3S_1) + \mathrm{H}_2$ was determined earlier,[16] if we exclude the diffusional correction term $\delta f_D$, apropos of which the following can be said.

In some of the absorption cells the quantities of He and Ne were commensurate, so that the rate of diffusion of the metastable atoms to the walls of the absorption cell could be significantly lower in the mixture than in the case of pure He. Consequently, the contribution of the diffusion process to the resonance line width should also be smaller in a He–Ne mixture. It is this factor that is taken into account in the expression (4). To estimate it, we used the diffusion coefficient values for He* in He, equal to $D_{\mathrm{He^*-He}} = 420 \pm 10\ \mathrm{cm^2\text{-}sec^{-1}}$ (Ref. 11) and He* in Ne, equal to $D_{\mathrm{He^*-Ne}} = 380\ \mathrm{cm^2\text{-}sec^{-1}}$ (when scaled to 1 mm Hg). The diffusion length was 0.9 cm, since we used $\varnothing 46 \times 80$ cylindrical absorption cells. The magnitude of the diffusional correction did not then exceed 0.2 kHz, and was significantly smaller than the resonance-line broadening $\Delta f_{\mathrm{Ne}}$, which varied between 1.4 and 3.4 kHz.

The value of the diffusion coefficient $D_{\mathrm{He^*-Ne}}$ was computed from the formula of Hirschfelder *et al.*[17] on the basis of an interaction potential for the He*–Ne system[18] that, in the region of interest to us, can be approximated by the power function $V^* = dR^{-n}$ with the parameters $d = 11$ and $n = 4.9$.

It should also be noted that the spin exchange between the $2^3S_1$ helium atoms and the neon atoms in the $2s_2$ and $2s_3$ states, as well as their mutual Penning ionization can be neglected. Although the magnitudes of the cross sections of these processes can be $\sim 10^{-14}\ \mathrm{cm}^2$, which is three orders of magnitude greater than the measured helium-deactivation cross sections, their contribution to the resonance line width does not exceed 0.1%. This is explained by the fact that the concentration of the Ne atoms in the $2s_2$ and $2s_3$ excited states under the conditions of the performed experiment is not higher than $10^{10}\ \mathrm{cm}^{-3}$, i.e., is six orders of magnitude lower than the concentration of the Ne atoms in the ground state.

TABLE II. Experimental data obtained in the investigation of the process of excitation transfer from He* to Ne by the method of optical orientation of atoms.

| $T$, K | $C$, $10^{-12}\ \mathrm{cm^3\text{-}sec^{-1}}$ | $\sigma$, Å$^2$ |
|---|---|---|
| 178 | 0.64±0.16 | 0.060±0.015 |
| 207 | 1.06±0.09 | 0.092±0.008 |
| 241 | 2.02±0.16 | 0.163±0.013 |
| 268 | 3.24±0.29 | 0.249±0.020 |
| 300 | 4.80±0.34 | 0.348±0.024 |

Thus, up to the readily estimable diffusion term, the cross section for excitation transfer from He* to Ne practically coincides with the cross section for destruction of the metastable He* state, which is due to the addition of neon. This guarantees the possibility in principle of determining to within a few percent the reaction rate of the process (1) on the basis of the relations (3) and (4).

In Table II we present the experimental data. The error in the determination of the quantity $C(T)$ is determined by both the errors in the measurement of the resonance-line broadening $\Delta f_{\mathrm{Ne}}$ and the inaccuracy in the determination of the neon pressure in the absorption cell, the latter factor being the more important one. The greater the contribution made by the process (1) to the resonance line width of He* is, and the more absorption cells with different Ne concentrations we use, the smaller is this error. The greatest relative error in the determination of the reaction rate of the process (1) occurs at $T = 178$ K.

## 3. DISCUSSION OF THE RESULTS

In Fig. 2 we show the temperature dependence, obtained in the present work, of the reaction rate of the process (1), as well as the data of other authors.[5,8] It can be noted that in the range from 178 to 300 K the reaction rate of the process (1) increases by a factor of 7.5, whereas, according to Jones *et al.*[5] and Arathoon,[8] the reaction rate increases by factors of 3.5 and 4, respectively. At $T = 300$ K the results of the present work are close to only the data presented in Refs. 2, 4, 5, and 10, and significantly differ from the data given in the other papers[1,8] (see Tables I and II).

The critical temperature dependence of the reaction rate of the process (1) is due to the essentially nonres-

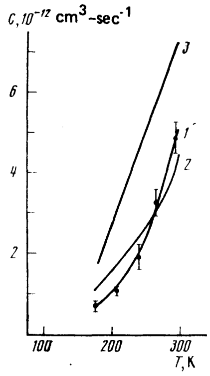

FIG. 2. Temperature dependence of the reaction rate of the process of excitation transfer from He* to Ne: 1) experimental data of the present work; 2) the data of Jones *et al.*[5]; 3) Arathoon's data.[8]

onance character of the process of excitation transfer from the $2^3S_1$ He level to the $2s_2$ and $2s_3$Ne levels, which are lower respectively by $\Delta E' = 0.039$ eV and $\Delta E'' = 0.058$ eV, values which are commensurate with the thermal energy $3/2\ kT$.

According to Refs. 18 and 19, the He*-Ne interaction potential for energy range investigated in the present work has a repulsive character, the He*-Ne collisions occurring at quite large internuclear distances $R$ (of the order of 7-9 a.u.). This enables us to use Nikitin's[12] simple exponential model, in which in the transition region between two states of the quasimolecule (HeNe)* the difference between the diagonal interaction matrix elements is assumed to be constant ($\varkappa$ = const), while the off-diagonal matrix element is approximated by a simple exponential function ($\Delta = Ae^{-\alpha R}$). According to Smirnov,[20] the cross section of the nonresonance process is given by the expression

$$\sigma = {}^1/_2 k\pi R_n{}^2 f(\gamma), \tag{5}$$

where $R_n$ (the center of the nonadiabaticity region) is found from the condition

$$\varkappa = \Delta, \tag{5a}$$

$$f(\gamma) = \int_0^1 \frac{2x\,dx}{\mathrm{ch}^2(\gamma/x)}, \quad \gamma = \frac{\pi}{2}\frac{\varkappa}{\alpha v}, \tag{5b}$$

and $k$ is a coefficient connected with the possibility of the reaction's being forbidden because of the law of conservation of angular momentum.

If we assume that the total angular momentum of the system is conserved, then $k = 1$. The experimental verification of this assumption is difficult, and falls outside the limits of the present work. We also assume that $\varkappa = \Delta E'$, $\Delta E''$, i.e., that the difference between the diagonal interaction matrix elements is equal to the resonance defect. It turns out that, for the investigated energy range, the dependence of the cross section on energy is, according to (5b), close to $\exp(-2.2\gamma)$. Since according to the data given in Table II, the cross section changes by almost a factor of 6 when the velocity of the relative motion changes by only a factor of 1.3, it is evident that the cross section should decrease by an even greater factor when the resonance defect is increased by a factor of 1.65 (i.e., when we go over from the $2s_2$ to the $2s_3$ level). From this it follows that the excitation transfer to the $2s_3$ level should form only an insiginificant fraction of the excitation transfer to the $2s_2$ level. This justifies the use of the two-level approximation in the analysis of the process of excitation transfer from He* to Ne.

Analysis of the experimental data in accordance with the formula (5) allowed the determination of the exchange interaction parameters: $A = 8\times 10^3$ and $\alpha = 1.38$ (in a.u.). These parameter values are highly rough estimates, on account of a number of assumptions made in the course of the derivation of (5) and the use of the corresponding model for the He*-Ne system. Nevertheless, on the basis of the values obtained for the parameters $A$ and $\alpha$, we can estimate the values of the cross sections and, consequently, the reaction rates of the process (1) outside the investigated temperature range; these values for $T > 300$ K are shown, in particular, in Fig. 3. It turned out also that the cross section of the process is only $10^{-19}$ cm$^2$ at $T = 77$ K. For the temperature range from 300 to 700 K the reaction-rate values computed for the process (1) from (5) with the use of the exchange-interaction parameters found in the present work are in agreement with the experimental results of Lindinger *et al.*[7]

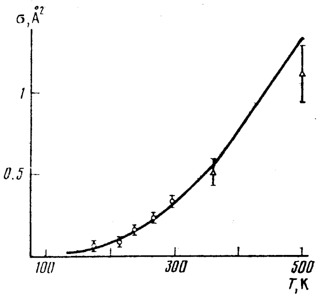

FIG. 3. Temperature dependence of the cross section for excitation transfer from He* to Ne. The solid curve was constructed from the formula (5) on the basis of an exchange potential having the form $\Delta = 8\times 10^3 \exp(-1.38R)$ in the transition region; c) experimental values obtained in the present work; $\Delta$) experimental values obtained by Lindinger *et al.*[7]

Thus, the use of the Rosen-Zener-Demkov model allows us to explain sufficiently well the temperature dependence of the reaction rate of the process of excitation transfer from He* to Ne.

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