Vol. 17(2), pp. 45-50, July-September, 2022
DOI: 10.5897/IJPS2022.5001
Article Number: 6FC6EBF69682
ISSN: 1992-1950

http://www.academicjournals.org/IJPS

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International Journal of Physical Sciences

Full Length Research Paper

Excitation of $^3G°$ levels of nickel atom by electron impact

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Received 25 July, 2022; Accepted 31 August, 2022

Excitation of $^3G°$ levels of the nickel atom was investigated using the method of extended atomic and electron crossing beams with registration of the optical signal of excited atoms. At an energy of exciting electrons of 50 eV, 14 excitation cross sections were measured. In the electron energy range of 0 to 200 eV, optical excitation functions (OEF) were recorded for transitions from five levels. Based on the results obtained, the total level excitation cross sections are calculated. All these results are received for the first time.

Key words: Nickel atom, cross section, optical excitation function (OEF), energy level, spectral line.

INTRODUCTION

Nickel, originally discovered by the Swedish metallurgist Axel Frederick Cronstedt in 1751, was not recognized as a metal by scientists for a long time. Only after the publication in 1804 by I. Richter of the article on obtaining "absolutely pure" nickel was finally recognized as a metal and by the beginning of the 20th century it had become extremely widespread and had numerous applications. At the same time, the content of nickel in the Earth's lithosphere is relatively low: its clarke is 0.008% (mass percent), which is approximately an order of magnitude greater than the clarkes of Nb, Th, Ge, Be and Sc (Wedepohl, 1967): 800 times less than the clarke of iron and 3.5 times more than the clarke of cobalt, its closest relatives in the table of elements.

Despite its relatively low abundance on the Earth's surface, nickel at the present time finds a huge number of applications due to a wide range of useful properties: Pure nickel is hard and ductile, resistant to high temperatures, exhibits the effect of magnetostriction, and has low vapor pressure, which is very important for vacuum technology. It is part of many alloys with unique properties: chromel, alumel, copel, nickel silver, cupronickel, monel metal, platinum, in short, several thousand alloys. Nickel complex compounds and catalysts based on it are no less important. Finally, the pinnacle of its originality is nickel carbonyl.

Research continues on the various properties of nickel. Along with research in the field of catalysis, the optical characteristics of free nickel atoms and ions present in the atmospheres of stars, as well as in the plasma of

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scientific and technological installations are also being studied. The need to obtain such data to improve the processes and apparatuses of modern semiconductor electronics was discussed in detail in review (Huo and Kim, 1999), as well as in recent work (Bartschat and Kushner, 2016). At the same time, the amount of such data can be large for each specific object. Thus, in the experimental study of the radiative characteristics of atoms, most modern publications contain the results of measurements of transition probabilities or branching factors for several hundred spectral lines. Experimental data on the cross sections for inelastic collisions of electrons with atoms are much less numerous and, moreover, have not yet been obtained for all atoms. In particular, for the nickel atom, the electron impact excitation cross sections for transitions from the ^3F (Smirnov, 2001), $^3F°$ (Smirnov, 2009), $^3P°$ and $^3D°$ (Smirnov, 2012) levels, as well as from the levels belonging to the $3d^94d$, $3d^95d$, $3d^96s$ (Smirnov, 2013) were obtained. In these works, the experiment was carried out by the method of extended crossed electron and atomic beams, a detailed discussion of which is given in recent papers (Smirnov, 2015; Smirnov, 2016).

Thus, the excitation by electron impact of all triplet levels of the nickel atom, except for $^3G°$, has been experimentally studied up to now, the excitation of which is the subject of this work.

EXPERIMENTAL

As in previous experiments with nickel, the method of extended crossing beams was used. Since the equipment and the technique for conducting the experiment by the method of extended crossing beams have recently been published in previous studies (Smirnov, 2015, 2016), here the study only indicate the basic conditions for conducting experiments with nickel.

Nickel with a purity of 99.99% was evaporated from a graphite cup-shaped crucible as a result of power supply to the surface of the nickel by an electron ray. The choice of graphite as the crucible material was determined by the fact that molten nickel intensively dissolves all metal crucible materials in contact with them. When graphite was used as the crucible material, the melting mode was chosen so that the melt zone did not come into contact with the graphite surface. Even a relatively slow dissolution of graphite would lead to contamination of the atomic beam with carbides.

Like the atoms of most metals, the nickel atom has a group of low-lying levels that can be populated as a result of thermal excitation when the metal evaporates. Since the measurement of the distribution of atoms over low-lying levels under the conditions of the experiment is an extremely difficult task, the population of these levels was estimated under the assumption that the Boltzmann distribution is valid. At the melt surface temperature (T = 1750 K), the following population values were obtained (in % of the total number of atoms in the beam; level energy are given in brackets in cm^{-1}): $3d^8(^3F)4s^2\,^3F_4$ (0) - 42.1; $3d^9(^2D)4s\,^3D_3$ (204) - 27.5; $3d^9(^2D)4s\,^3D_2$ (879) - 11.2; $3d^8(^3F)4s^2\,^3F_3$ (1332) - 10.8; $3d^9(^2D)4s\,^3D_1$ (1713) - 3.36; $3d^8(^3F)4s^2\,^3F_2$ (2216) - 3.66; $3d^9(^2D)4s\,^1D_2$ (3409) - 1.38. Thus, populations of more than one percent have levels of three terms: $3d^8(^3F)4s^2\,^3F$; $3d^9(^2D)4s\,^3D$; $3d^9(^2D)4s\,^1D$. At the same time, the contribution of each of the low-lying levels to the population of any upper level can be determined only theoretically.

The concentration of atoms in the intersection zone of an atomic beam with an electron beam was 1.0 × 10^{10} cm^{-3} in the main study regime and decreased to 1 × 10^9 cm^{-3} when studying resonant transitions in order to minimize reabsorption. In the whole operating energy range of 0 to 200 eV the current density of electron beam did not exceed 1.0 mA/cm^2. The spectral resolution of the setup was about 0.10 nm, and only for the most intense lines could it be improved to 0.05 nm. The measurement error of the relative values of the cross sections, depending on the intensity of the line and its position in the spectrum, was within 3 to 10%; the absolute values of the cross sections were determined with an error of ±20 to 27%.

RESULTS AND DISCUSSION

The optical emission spectrum resulting owing to the bombardment of nickel atoms by electrons with energy of 50 eV was recorded in the wavelength range of 220 to 622 nm. Of almost 300 spectral lines found on the spectrogram, more than 90% belong to the NiI spectrum; the NiII spectrum (excitation with simultaneous single ionization) is excited much less efficiently. Of the 260 NiI lines, sixteen result owing to transitions from $^3G°$. For transitions from five levels, the dependence of the cross section on the energy of the exciting electrons (optical excitation function, OEF) was recorded.

Nickel is the last element in the VIII group of the IV period, that is, in the iron group, having in the ground state of the atom a set of filled electron shells $1s^22s^22p^63s^23p^63d^84s^2$, with the exception of the penultimate, which contains 8 $3d$-electrons, but not 10. Therefore, the system of energy levels of the nickel atom contains a set of low-lying even configurations $(3d + 4s)^{10}$ and is qualified in Litzen et al. (1993) as one of the simplest. In Litzen et al. (1993), information about four odd triplet G-terms is given (two of them are not filled in); even ^3G terms are absent. But at the same time, it should be noted that the large majority of the even levels of the nickel atom are classified on the basis of the J_cK-bond, which is more adequate for these levels, and not in the notation of the LS-bond. The location of the levels studied in this work and the transitions between them are clearly shown in Figure 2. It can be seen that the possibilities of allowed transitions from the $^3G°$, as well as branching, are very limited in the case of the nickel atom.

The format of Figure 1 is generally accepted for the representation of atomic OEF; the scale along the abscissa axis is logarithmic, along the ordinate axis is linear. All curves are normalized by one at the maximum. Each curve has an individual zero reference level along the ordinate axis. The offset along the ordinate axis is set in such a way as to avoid intersecting or touching the curves.

The results of measuring the excitation cross sections with the addition of the necessary reference spectroscopic data are presented in Table 1. Here, the wavelength λ, the transition according to (Litzen et al.,1993), the values

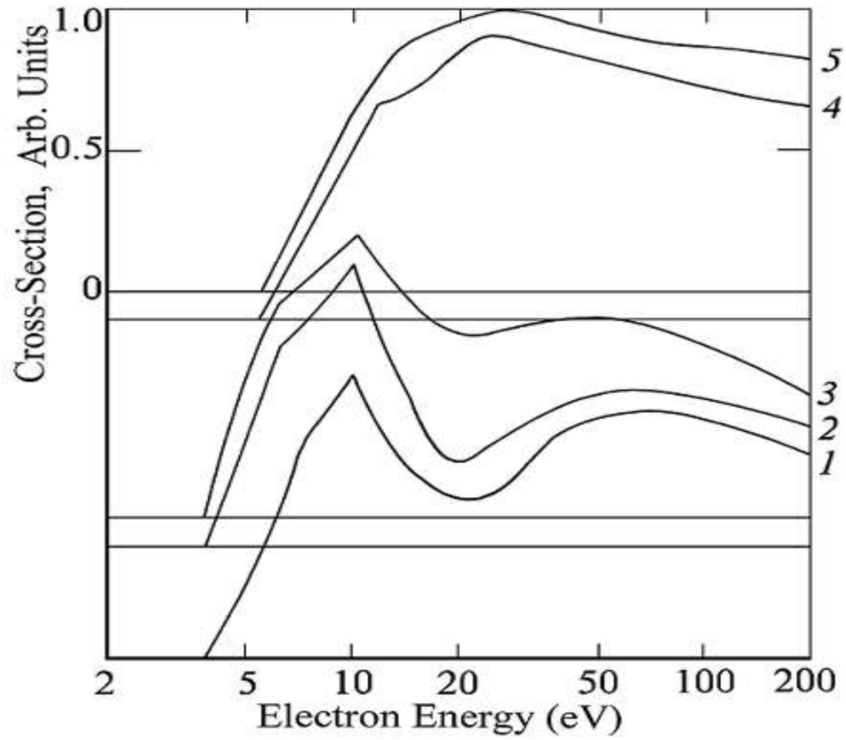

Figure 1. Optical excitation functions of transitions from the $^3G°$ levels of the nickel atom.

of the internal quantum numbers of the lower J_{low} and upper J_{up} levels, and the energies of the lower E_{low} and upper E_{up} levels are given. The Q_{50} column shows the values of the cross sections at the energy of exciting electrons of 50 eV, and the Q_{max} column shows the cross sections in the maximum of OEF. The position of the maximum is indicated in column $E(Q_{max})$. The numbers in the OEF column correspond to the numbering of the curves in Figure 1. All reference spectroscopic data are given according to Litzen et al. (1993).

As is known, all experimental methods for measuring collision cross sections based on detection of optical radiation of excited atoms make it possible to directly determine the cross sections for excitation of spectral lines Q_{ki} corresponding to spontaneous transitions from level k to level i. However, the main characteristic of inelastic electron-atom collisions, which is mainly used in the collisions theory and in solving practical problems, is the excitation cross section q_k of the energy level k. The relation between these quantities is established by the known relation:

$$q_k = \sum_i Q_{ki} - \sum_l Q_{lk} \quad (1)$$

where the left sum is the total excitation cross section of level k (taking into account both the direct excitation of the level k by electron impact from the initial state and its population by cascade transitions from higher levels), and the right sum is the total cross section for the population of level k by cascading transitions.

Since the optical signal in experiments with intersecting beams is significantly limited due to the low target density(necessary upper restrictions on the values of the atomic concentration and electron current density in the beams), as a rule, only a small number of terms are recorded in both the left and right sums. In modern studies, some basic values of atomic constants (cross sections, lifetimes) are measured in intersecting beams, and the range of transitions under study is expanded by using the branching factors BR obtained in a gas-discharge experiment. Since:

$$\sum_i Q_{ki} = Q_{ki}/BR_{ki} \quad (2)$$

Part of the level designations, as far as possible, is removed from the field of the figure under the abscissa axis. The numbers in the figure field to the right of the levels are the values of J. Vertical dashed lines separate states that differ in parity.

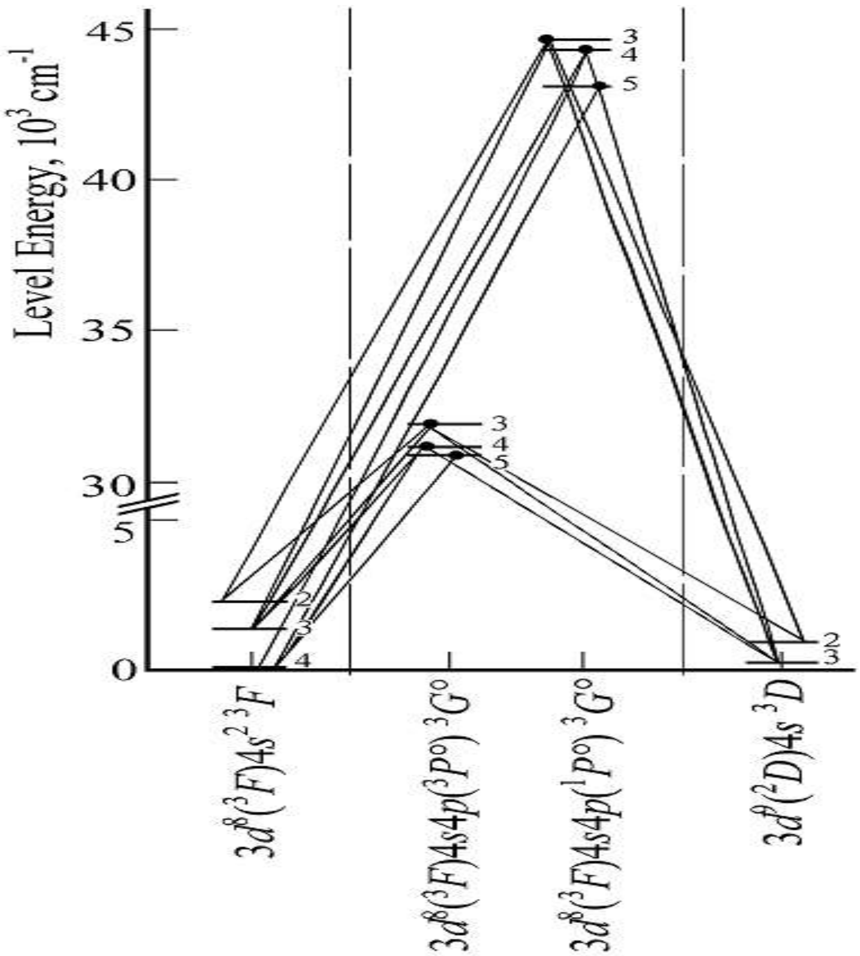

Figure 2. Partial states diagram of nickel atom with transitions studied.

Table 1. Excitation cross-sections of NiI spectral lines (transition from $^3G°$ levels).

λ, nm	Transition	J_{low}-J_{up}	E_{low}, cm^{-1}	E_{up}, cm^{-1}	Q_{50}, 10^{-18} cm^2	Q_{max}, 10^{-18} cm^2	$E(Q_{max})$, eV	OEF
225.588	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	4-4	0	44314	0.21	0.23	25	4
226.635	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	3-4	204	44314	0.29	0.32	25	4
228.840	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	2-3	879	44565	0.45	-	-	-
231.234	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	3-3	1332	44565	24.3	-	-	-
232.003	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	4-5	0	43089	64.1	70.0	28	5
232.580	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	3-4	1332	44314	40.9	45.0	25	4
236.064	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^1P°)\ ^3G°$	2-3	2216	44565	0.78	-	-	-
316.551	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	3-3	204	31786	0.08	0.09	10	1
322.698	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	4-4	0	30979	0.67	1.26	10	2
323.293	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	4-5	0	30922	17.2	24.5	10	3
323.465	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	2-3	879	31786	2.41	2.83	10	1
324.846	$3d^9(^2D)4s\ ^3D$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	3-4	204	30979	1.83	3.44	10	2
328.270	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	3-3	1332	31786	0.54	0.64	10	1
337.199	$3d^8(^3F)4s^2\ ^3F$-$3d^8(^3F)4s4p(^3P°)\ ^3G°$	3-4	1332	30979	9.15	17.2	10	2

Table 2. Total excitation cross-sections of $^3G°$ energy levels of nickel atom.

Term	E_k, cm^{-1}	J_k	$\Sigma Q_{50,\,10}{}^{-18}$ cm^2	ΣA_{ki}, 10^6 c^{-1}	ΣA_{ki} (total), 10^6 c^{-1}	ΣA_{ki} (total)/ΣA_{ki}	ΣQ_{50} (total), $_{10}{}^{-18}$cm^2
$3d^8(^3F)4s4p(^3P°)$ $^3G°$	30922	5	17.2	5.49	5.49	1.00	17.2
	30979	4	11.65	3.277	3.277	1.00	11.65
	31786	3	3.03	2.472	5.179	2.09	6.35
$3d^8(^3F)4s4p(^1P°)$ $^3G°$	43089	5	64.1	-	-	-	64.1
	44314	4	41.4	345.02	345.02	1.00	41.4
	44565	3	26.4	483.7	499.16	1.033	27.3

To determine the total cross section, it suffices to measure the excitation cross section of at least one transition from the level under study and the corresponding branching factor. However, both Q_{ki} and BR_{ki} have individual errors, so to improve the accuracy of the calculating the total excitation cross section of level k is advisable to use all available information about Q_{ki} and BR_{ki}.

To obtain BR, it is necessary to have a set of the radiative transition probabilities A_{ki} from level k (or oscillator strengths), since $Q_{ki}/Q_{km} = A_{ki}/A_{km}$. Detailed information about the A_{ki} of the nickel atom is contained in Wood et al. (2014). The calculation of the total excitation cross sections of the nickel atom energy levels is carried out in the present work on the basis of Table 1 of this paper and the results of Wood et al. (2014). It should be noted that transitions in the IR part of the spectrum are not taken into account in the calculation, since the results in Wood et al. (2014) are presented in the wavelength range λ = 212-779 nm.

The calculation results are presented in Table 2, where, in addition to the values previously deciphered in the text, are given: ΣQ_{50} is the sum of the cross sections for excitation of transitions from levels k, studied in this work (Bartschat and Kushner, 2016). Electron collision with atoms, ions, molecules, and surfaces: energy of exciting electrons of 50 eV; ΣA_{ki} is the sum of the probabilities of the same transitions from levels k; ΣA_{ki} (total) is the sum of the transition probabilities of all lines from levels k known from the data of Bartschat and Kushner (2016); ΣQ_{50} (total) is the total excitation cross section of level k (taking into account both the direct excitation of the level by electron impact from the initial state and its population by cascade transitions from higher-lying levels).

From Table 2 it can be seen that for levels $^3G°_J$ with J = 4 and 5, the correction factor ΣA_{ki} (total)/ΣA_{ki} is equal to one. This is an obvious result considering that only two even levels with J = 4 have less energy than the $^3G°$ levels, one of which is the main level, and the second is $3d^8(^1G)4s^2$ $^1G_4$ with energy E = 22102 cm^{-1}. For $^3G°$ levels with J = 5, transitions to all other low-lying even levels are strictly forbidden by the selection rule for ΔJ. For levels $^3G°$ with J = 3, along with completely allowed resonant transitions, there are transitions to levels of the low-lying triplet term $3d^9(^2D)4s$ ^3D (transition $^3G \rightarrow {}^3D$), forbidden by the selection rule for ΔL, however, this forbid not as strict as the forbid on ΔJ. The resulting transitions are shown in Figure 2 on the right side.

In the Table 2 in columns ΣA_{ki} and ΣA_{ki} (total) for the 43089 cm^{-1} level dashes are given, since the only completely allowed resonant transition is $3d^8(^3F)4s^2$ $^3F_4$ - $3d^8(^3F)4s4p(^1P°)$ $^3G°_5$, known from a number of works as an intense line, is absent in Wood et al. (2014) for an unknown reason. In addition, the 225.357 nm line (transition $3d^9(^2D)4s$ $^3D_3$-$3d^8(^3F)4s4p(^1P°)$ $^3G°_3$) was not included in Table 1, blended by a more intense nickel ion line, and the line at 338.088 nm is a rather intense transition $3d^8(^3F)4s^2$ $^3F_2$-$3d^8(^3F)4s4p(^3P°)$ $^3G°_3$, but blended with a much more intense line λ = 338,057 nm, since in both these cases the spectral resolution of our equipment was insufficient to resolve the blends.

It should be noted that, as can be seen from the results of Table 2, the excitation cross sections for the levels above the $^3G°$ term are, on average, greater than the cross sections below the one located approximately four times. One possible explanation for this fact is that the excitation of the lower term occurs mainly from the levels of the ground term in the process $3d^8(^3F)4s^2$ $^3F_J \rightarrow$ $3d^8(^3F)4s4p(^3P°)$ $^3G°_J$, while above the one located, in the process $3d^9(^2D)4s$ $^3D_J \rightarrow 3d^8(^3F)4s4p(^1P°)$ $^3G°_J$, that is, from the levels of the low-lying metastable term $3d^9(^2D)4s$ ^3D, the population of which is only 1.34 times less than that of the main term. To clarify this issue, theoretical consideration is needed.

As shown in Equation (1), to determine the excitation cross section of k level, it is necessary to subtract the contribution of the cascade population from the excitation cross section total level. Until now, this problem has remained unsolvable for experiment, since it requires summing the excitation cross sections of a huge number of low-intensity lines, most of them located in the IR range. The current situation can be illustrated by the example of the present work: earlier in Smirnov (2001), the excitation ^3F level of the nickel atom was studied, transitions from which provide cascade population of low-lying levels, including the $^3G°$ levels. In Smirnov (2001), 40 transitions from ^3F levels were registered, but only four of them end at the $^3G°$ levels, and three of these four end at the level E = 30922 cm^{-1} and only one at the level E = 30979 cm^{-1}.

Conclusion

Excitation $^3G°$ levels of the nickel atom by electron impact have been studied for the first time. The results obtained can be used for calculations and evaluations in problems of plasma physics, spectral diagnostics, etc.

CONFLICT OF INTERESTS

The author has not declared any conflict of interests.

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