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THE *KLL* AUGER SPECTRUM OF NEON FROM THE EC DECAY OF ^{22}Na

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KLL-оже спектр неона из радиоактивного распада ^{22}Na

С помощью электростатического бета-спектрометра при инструментальном разрешении 4 эВ исследован *KLL*-спектр оже-электронов неона, возникающих при электронном захвате в ^{22}Na. Это первое экспериментальное исследование *KLL*-спектра неона, возникающего в твердотельном окружении. Определены энергии и относительные интенсивности всех компонентов спектра. Измеренное значение абсолютной энергии доминирующего $KL_2L_3(^1D)$-перехода составило 824,5(19) эВ, что на 20 эВ больше, чем эти же значения, полученные в экспериментах со свободными атомами неона. Влияние твердотельных эффектов на относительные интенсивности *KLL*-переходов неона в пределах погрешностей измерений не выявлено.

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The *KLL* Auger Spectrum of Neon from the EC Decay of ^{22}Na

The low energy *KLL* Auger electrons of neon emitted after *EC* decay of ^{22}Na have been investigated with the 4 eV instrumental resolution using an electrostatic spectrometer and a solid state radioactive source. This is the first experimental investigation of the *KLL* Auger spectrum of neon originating in the solid state surrounding. Relative intensities and energies of all resolved spectrum components were determined. Measured absolute energy of the dominant $KL_2L_3(^1D)$ transition was found to be 824.5(19) eV, i.e. by about 20 eV higher than that obtained in experiments with free Ne atoms. Within the experimental uncertainties, no influence of solid state effects on relative intensities of the *KLL* transitions was found.

The investigation has been performed at the Dzhelepov Laboratory of Nuclear Physics, JINR.

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INTRODUCTION

The study of Auger transitions in atoms with closed atomic shells is very important for better understanding of Auger effect. The reason lies in reduced complexity of verification of influence of various correlation effects on transition energies and intensities. For the low Z region, neon ($Z = 10$) and argon ($Z = 18$) are suitable candidates. Neon is the heaviest atom in which the only *KLL* Auger transitions are possible.

Theoretical probabilities of the *KLL* transitions in Ne are available from calculations [1–3] performed in the framework of the *LS* scheme. In the work [1], calculations are based on the nonrelativistic Hartree–Fock–Slater model incorporating interaction between the $1s^2 2s^0 2p^6 - {}^1S_0$ and $1s^2 2s^2 2p^4 - {}^1S_0$ final states. H. Kelly [2] used Hartree–Fock approximation taking into account configuration coupling and correlation effects within the many-body perturbation theory. G. Howat et al. [3] detailed calculations with consideration of mixing between the final ${}^2S$ channels and with including of both relaxation and configuration-mixing effects.

Experimentally, the *KLL* Auger spectrum of neon was so far investigated only by external excitation of free atoms with photons or accelerated charged particles [4–7]. In the work [7], the *KLL* spectrum of Ne was induced by impact between Ne atoms and energetic heavy ions of H^+, Ne^{3+}, Ne^{10+}, Ar^{6+} and Ar^{10+}. The measured spectra were badly mixed with satellite lines arising from simultaneous ionization of internal and external shells of targeted atoms, as well as from other collision effects. According to the obtained experimental data, in general, energies and intensities of Auger transitions are independent of energy or type of incident particles. Exceptions revealed in [7] are Ar^{6+} and Ne^{3+} for which deviations of measured and predicted intensities of the $KL_1L_{2,3}(^3P)$ and $KL_2L_3(^1D)$ transitions reach the factor of two.

High sensitivity of probabilities of Auger transitions to details of wave functions indicates a possibility of influence of physical and chemical surrounding of atoms on energies and intensities of Auger lines [8]. It is obvious that in the case of transitions involving inner atomic shells, e.g., the *KLL* ones, such an influence will be more apparent when these shells lie close to the valence band. Experimental investigations of influence of physical and chemical surrounding on absolute energies and intensities of Auger transitions in light nuclei have been performed, e.g., in works [9–15]. The experimental data clearly show a relationship between the Auger transition energies and chemical and phase state of the atom. It has been found that chemical shift has a value of a few eV [9, 10]. The difference of absolute transition energies between gas and solid phase is significantly larger

and for light atoms it reaches a value from 12 to 16 eV [9, 11]. The influence of physical and chemical surrounding on relative intensities of Auger transitions was found to be less evident. In the above-cited works [12–15], there are unconvincing indications of such an influence. Relative experimental uncertainties in these works reached 25% for solid state samples. However, there are objective difficulties which prevent one to improve the experimental accuracy in determination of intensities of electron lines in low energy Auger spectra emitted by solid state samples. In the first place among them there is a background due to inelastically scattered electrons.

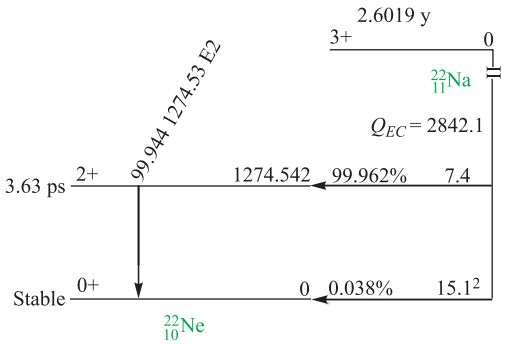

Fig. 1. The decay scheme of ^{22}Na [16]

In this paper, we present the results of the first experimental investigation of the *KLL* Auger spectrum of ^{22}Ne originating in the solid state radioactive sample after the electron capture decay of ^{22}Na ($T_{1/2} = 2.6$ y), see Fig. 1.

1. EXPERIMENTAL

1.1. Source preparation. The ^{22}Na was produced in the reaction ^{27}Al $(p, 3n3p)^{22}$Na. Aluminum was irradiated by 85 MeV protons at the phasotron of the JINR in Dubna. A thin source for electron spectroscopy was prepared by thermic evaporation of ^{22}Na in a vacuum on a carbon backing. Activity of the source was 14 μCi. Exact chemical state of the deposited microquantity of ^{22}Na on surface of the carbon backing is unknown. Most likely, it is Na_2O or chemical compounds like $NaHCO_3$, Na_2CO_3, NaOH. In any case, after preparation the source was transferred to the spectrometer on air, thus, the evaporated layer was in contact with oxygen. So, in all possible combinations, ions of Na were bounded with oxygen and have oxidation number $+1$.

1.2. Spectrum measurement and evaluation. The electron spectrum was analyzed using the electrostatic spectrometer [17], consisting of a retardation sphere followed by a double-pass cylindrical mirror energy analyzer. The spectrum was scanned under the constant instrumental resolutions of 4 eV with the step of 1 eV by changing the retardation voltage.

To decompose the measured spectrum into components a special computer code was used. The individual spectrum line shape was described by a convolution of Gaussian (the spectrometer response function) and «a model» function, which described the natural distribution of originating *KLL* Auger electrons in Ne atoms (Lorentzian) and its deformation due to inelastic scattering of the studied electrons in the source material. Systematic uncertainties of the description were estimated by the Monte Carlo method. For each Auger line, its position, height and natural width were fitted parameters. One of the measured *KLL* Auger spectra of ^{22}Ne with the results of its evaluation is shown in Fig. 2.

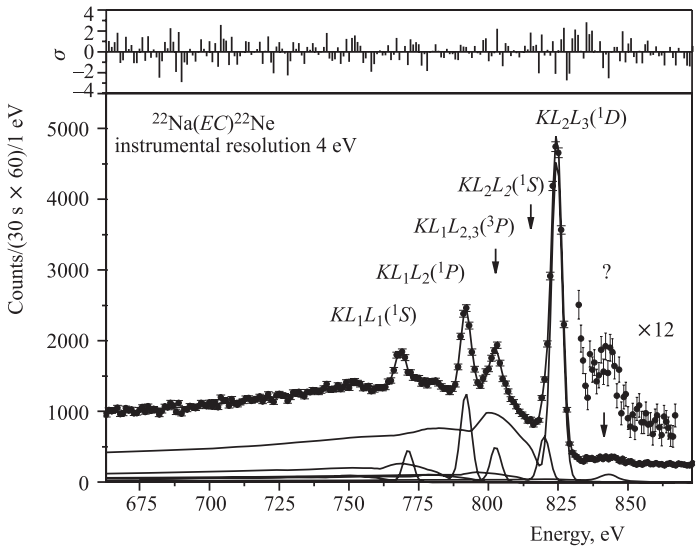

Fig. 2. An example of the experimental *KLL* Auger spectrum of ^{22}Ne from the EC decay of ^{22}Na

2. RESULTS AND DISCUSSION

2.1. Energies. In Table 1, our absolute energy of the dominant $KL_2L_3(^1D)$ transition is compared with the data available from the previous experimental and

theoretical works. As can be seen from the table, our value is higher by about 20 eV than the results of both the theory [2] and the measurements [6, 7] with free neon atoms.

Table 1. The absolute energy of the $KL_2L_3(^1D)$ Auger transition in Ne

Transition	Absolute energy, eV			
	Theory	Experiment		
	[2] LS(CIEC)[a]	[6]	[7]	This work
$KL_2L_3(^1D)$	804.51[b]	804.2(3)[b]	804.5(1)[b]	824.5(19)[c]

[a] Calculations in *LS* coupling with configuration interaction and electron correlation effects

[b] Related to vacuum level

[c] Related to Fermi level

It is difficult to point to a single reason of the disagreement found. Possible candidates can be: *i*) a difference in the phase state of Ne used in the measurements (free atoms [6, 7], solid state ^{22}Na source — our work), *ii*) an effect due to the origin of primary vacancies [18] (external excitation [6, 7] and the electron capture — the present work), *iii*) chemical effects. The «effect of atomic structure» – *ii*) (the presence of $3s$ electron in the atomic cloud of Ne in time of Auger processes after decay of ^{22}Na) influences substantially electron binding energies and can be the dominant one. Nevertheless, it is difficult to assign a rate for each of the listed effects, since: i) the exact chemical form of ^{22}Na atoms in our source is unknown; *ii*) to our knowledge, information on Auger transition energies for the solid phase is unavailable; *iii*) theoretical and experimental knowledge on influence of the «atomic structure effect» on the Auger transition energies is quite poor. Still now, there are only a few works [19, 20] in which an energy shift due to the «atomic structure effect» has been observed for the $KL_2L_3(^1D)$ and $L_3M_{4,5}\,M_{4,5}(^1G_4)$ Auger transitions for elements with $Z = 26 \div 70$. In these works, energy shifts (experiment–theory) were found to have values from $+6$ to $+27$ eV. Attempts of theoretical estimations of influence of the «atomic structure effect» on the energy of the $KL_1L_1(^1S_0)$ transition were performed in works [21, 22] for Ag, Mn, Eu, Tm and W generated in the *EC* decays of Cd, Fe, Gd, Yb and Re, respectively. Since the contribution of the physicochemical shift to the total Auger energy shift is a mix-up with the shift due to the «atomic structure effect» there is no ultimate validation of the theoretical predictions.

Our relative energies of the *KLL* transitions in ^{22}Ne are in good agreement with both the theory [2] and the experimental results [6, 7], with the exception of

the $KL_2L_2(^1S)$ transition, for which our value (and that of [7]) is significantly higher than the theoretical prediction [2] (see Table 2).

Table 2. Relative energies of the *KLL* Auger transitions in Ne

Transition	Relative energies $(KL_iL_j(^1S) - KL_2L_3(^1D))$, eV			
	Theory	Experiment		
	[2] LS(CIEC)[a)]	[6]	[7]	This work
$KL_1L_1(^1S)$	–56.36	–54.3(3)	–56.1(1)	–56.0(2)
$KL_1L_2(^1P)$	–32.8	–32.9(3)	–32.6(1)	–32.7(1)
$KL_1L_{2,3}(^3P)$	–22.06	–22.1(3)	–22.3(1)	–21.9(2)
$KL_2L_2(^1S)$	–3.24	–3.6(3)	–3.8(1)	–4.3(5)
$KL_2L_3(^1D)$	0	0	0	0
Unknown	—	—	—	+ 18.5(5)

[a)] Calculations in *LS* coupling with configuration interaction and electron correlation effects

2.2. Intensities.

In Table 3, our relative intensities of the *KLL* transitions in Ne are compared with the predictions [1–3] and with the experimental data [4–7] obtained for free neon atoms.

Table 3. Relative intensities of the *KLL* Auger transitions in Ne

Transition	Intensity $(KL_iL_j/\sum KLL)$, %					
	Theory			Experiment		
	[1] LS(CI)[a)]	[2] LS(CIEC)[b)]	[3] LS(CIRCM)[c)]	[7]	[4–6]	This work
$KL_1L_1(^1S)$	7.0	5.8	5.0	5.9(3)	6.1(1)	6.0(4)
$KL_1L_2(^1P)$	20.9	16.5	16.7	17.3(3)	16.9(7)	16.3(6)
$KL_1L_{2,3}(^3P)$	6.9	5.9	7.8	6.3(3)	6.2(5)	6.6(5)
$KL_2L_2(^1S)$	6.9	10.2	9.2	9.7(1)	10.0(7)	8.3(10)
$KL_2L_3(^1D)$	58.2	61.5	61.3	60.9(8)	61.6(3)	60.0(12)
Unknown	—	—	—	—	—	2.7(5)

[a)] Calculations in *LS* coupling with configuration interaction

[b)] Calculations in *LS* coupling with configuration interaction and electron correlation effects

[c)] Calculations in *LS* coupling with incorporated relaxation and final-state channel mixing

It is seen from the table that probabilities of the $KL_1L_1(^1S)$, $KL_1L_2(^1P)$ and $KL_1L_{2,3}(^3P)$ transitions are significantly suppressed in more complex calculations

[2, 3] incorporating correlation and relaxation effects in comparison with «more simple» ones [1]. Exception is the $KL_1L_{2,3}(^3P)$ transition for which the influence of the relaxation effects leads to the increase of its probability. Our experimental intensities of the *KLL* transitions in Ne, as well as those from works [4–7], have better agreement with calculations of H. Kelly [2]. Agreement of our results with the data [4–7] obtained for free atoms of neon shows that influence of the solid state effects on the *KLL* transition intensities is smaller than the achieved experimental uncertainties.

2.3. Electrons with energies above main *KLL* Auger group. As can be seen in Fig. 1, our experimental spectrum exhibits clear excess of events above the background at the energy higher by about 18 eV than the energy of the $KL_2L_3(^1D)$ line. We found that the intensity of this «bump» is in the direct correlation with the intensity of the *KLL* Auger electrons of ^{22}Ne. Gamma spectroscopy of the measured ^{22}Ne source did not reveal any contamination by other isotopes. Thus, it seems evident that electrons in the bump originated in the decay of ^{22}Na. The nature of these electrons is not obvious.

CONCLUSION

Using the advantage of high resolution electron spectroscopy with the electrostatic spectrometer, the first experimental investigation of the *KLL* Auger spectrum of neon from the EC decay of ^{22}Na in the solid state source was performed. The measured absolute energy of the main $KL_2L_3(^1D)$ transition was found to be 824.5(19) eV, i.e. by about 20 eV higher than that obtained in experiments with free Ne atoms. Thus, an influence of the solid state effect and «the atomic structure effect» in neon atom were revealed for the first time. Relative intensities and energies of all resolved components of the *KLL* Auger spectrum of neon were also determined. Influence of the solid state and other effects on intensities of the Ne *KLL* Auger lines in the case of the ^{22}Na solid state source was found to be smaller than the achieved experimental uncertainties.

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