THERMAL CONDUCTIVITY OF ARGON AND ARGON – NEON MIXTURES AT HIGH TEMPERATURE

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UDC 536.23:536.45

The thermal conductivity of neon and argon–neon mixtures is studied in the temperature range 400-1500°K. This is the first recording of such data above 793°K.

Present-day vacuum, electronic, plasma, and welding techniques, as well as the metallurgy of high-melting-point metals and their alloys, make wide use of monatomic inert gases and their mixtures. Thus, a knowledge of the thermophysical properties of these gases and mixtures is required, especially the thermal-conductivity coefficient, over a wide parameter range [1].

The thermal conductivity of neon and its mixtures with argon has been studied in a device with a molybdenum measurement cell [2], which was used previously to measure the thermal conductivity of helium, argon, and their mixtures in the temperature range 400-1500°K [2-4]. In [2-4] a detailed analysis was made of various factors distorting the true value of the measured thermal conductivity, such as thermal radiation, temperature changes, free convection, heat loss from end faces, temperature differential within the molybdenum tube wall, eccentricity, and thermal diffusion. It has been shown [4] that the greatest effect at high temperatures is produced by radiation and temperature shift.

Heat transferred by radiation was considered by using the Stefan–Boltzmann equation with experimental data on the temperature dependence of the integral hemispherical emissivity of the measurement wire, as obtained in special experiments at All-Union Institute of Aviation Materials (VIAM) [5]. In our experiments the contribution of radiation to the total thermal flux increased with increase in temperature and reached its highest value (of the order of 27%) in the case of a mixture with high concentration of the heavy component (0.2 Ne-0.8 Ar) at T = 1474°K. With increase in the mixture of light component concentration the fraction of radiation decreased, comprising about 17% for pure neon.

The correction for temperature shift was determined experimentally [2]. At a temperature of 373°K for neon and argon–neon mixtures this correction did not exceed 1%. With increase in temperature it increases, reaching 13% at T = 1503°K.

With increase in the mixture of heavy component concentration the correction for temperature shift decreases (for a mixture 0.2 Ne–0.8 Ar it does not exceed 3% at T = 1474°K).

Measurement data and the experimentally determined thermal-conductivity values are presented in Table 1. The maximum error of the measured thermal-conductivity values was evaluated by the method of [6]. This maximum value for an Ne–Ar mixture in the temperature range 400-1500°K does not exceed ± 2-4%.

The thermal conductivity of neon (99.882%) containing not more than 0.001% hydrogen, 0.1% helium, 0.001% oxygen, 0.01% nitrogen, and 0.02 g/m^3 moisture was studied over the temperature range 373-1503°K with the aid of the experimental cell of [4].

Figure 1 compares the present experimental data on thermal conductivity of neon with recommended values [7, 8], the experimental data of [9-11], and theoretical values calculated with strict molecular-kinetic theory [23] using various intermolecular interaction potential models with corresponding potential parameters (Table 2).

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Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 33, No. 5, pp. 848-856, November, 1977. Original article submitted March 11, 1977.

0022-0841/77/3305-1318$07.50

TABLE 1. Measurement Data and Experimental Values of Thermal Conductivity of Argon–Neon Mixtures and Neon at Atmospheric Pressure, I_0 = 0.002 A

ΔT_g, °K	$\bar{T}$, °K	$Q \cdot 10^6$, W	$Q_r \cdot 10^6$, W	$Q_t \cdot 10^6$, W	$\lambda \cdot 10^3$, W/m·°K	ST_{sh}, %
			0,2 Ne—0,8 Ar			
9,26	392	22951	100	22851	26,6	0,3
9,22	505	28251	284	27967	32,7	0,45
9,19	597	31463	553	30910	36,3	0,55
9,15	658	33923	816	33107	39,0	0,7
9,06	816	39649	1926	37723	44,9	1,0
9,02	896	43966	2793	41173	49,2	1,1
8,9	991	48072	4147	43925	53,2	1,5
8,81	1071	49828	5613	44215	54,1	1,6
8,75	1122	53252	6743	46509	57,3	1,7
8,65	1227	57405	9580	47825	59,6	2,1
8,55	1320	62754	12785	49969	63,0	2,4
8,35	1474	72012	19570	52442	67,7	2,9
			0,4 Ne—0,6 Ar			
10,86	398	33572	125	33447	33,2	0,4
10,76	490	39123	291	38832	38,9	0,7
10,68	576	42962	560	42402	42,8	0,9
10,6	654	48527	954	47593	48,4	1,2
10,48	753	52269	1620	50649	52,1	1,5
10,37	859	58532	2733	55799	58,0	1,8
10,26	978	65701	4597	61104	64,2	2,2
10,18	1049	68202	6064	62138	65,8	2,7
10,11	1145	74514	8580	65934	70,3	3,0
9,98	1259	80435	12478	67957	73,4	3,6
9,84	1376	89021	17636	71385	78,2	4,2
9,72	1501	98884	24940	73944	82,0	4,7
			0,6 Ne—0,4 Ar			
13,73	394	51103	155	50942	40,0	0,5
13,66	474	57984	325	57659	45,5	0,9
13,56	567	65332	671	64661	51,4	1,4
13,47	647	72581	1136	71445	57,2	1,7
13,25	779	83093	2456	80637	65,6	2,5
13,13	877	89479	3847	85632	70,3	3,0
13,0	998	99373	6388	92985	77,1	3,6
12,85	1108	107700	9722	97978	82,2	4,4
12,7	1232	118453	14786	103667	88,0	5,1
12,5	1347	128202	21050	107152	92,4	6,0
12,3	1499	141587	32047	109540	96,0	7,0
			0,8 Ne—0,2 Ar			
12,98	399	58991	155	58836	48,9	0,9
12,61	496	68209	359	67850	58,0	1,5
12,32	592	73643	725	72918	63,8	2,2
15,26	643	80057	1011	79046	65,9	2,4
11,94	805	90983	2480	88503	79,9	3,7
11,75	910	100830	4032	96798	88,8	4,8
11,64	1030	109665	6649	103016	95,4	5,4
11,5	1101	114258	8635	105623	99,0	6,1
11,4	1166	119742	10817	108925	103,0	6,7
11,3	1247	127339	14125	113214	108,0	7,3
11,1	1369	135909	20582	115327	112,0	8,5
10,91	1517	151380	30936	120444	119,0	9,8
			Ne			
12,8	373	67345	115	67230	56,9	0,8
12,58	475	78688	307	78381	67,5	1,7
12,73	581	89184	689	88494	77,5	2,8
12,23	693	99955	1396	98559	87,3	3,7
12,05	781	108939	2259	105780	95,1	4,7
11,91	875	116354	3556	112798	102,6	5,5
11,77	958	124655	5139	119516	110,0	6,4
11,47	1113	137412	9292	128120	121,0	8,2
11,33	1215	145995	13169	132826	127,0	9,0
11,07	1341	158447	19466	138981	136,0	10,0
10,91	1422	166641	24633	142008	141,0	11,7
10,71	1503	174946	30602	144344	146,0	12,9

TABLE 2. Potential Function Parameters for Interaction of Homogeneous and Inhomogeneous Molecules

Mixture	Potential function	Variant	ε_1/k, °K	ε_2/k, °K	ε_{12}/k, °K	σ_1, Å	σ_2, Å	σ_{12}, Å	α_1	α_2	α_{12}	Reference
Ne—Ar	(12—6)	I	35,7	124	66,535	2,789	3,418	3,1035	—	—	—	[23]
	(12—6)	II	27,5	116,0	56,48	2,858	3,465	3,1615	—	—	—	[23]
	(12—6)	III	43	135	76,191	2,73	3,36	3,045	—	—	—	[24]
	(12—6)	IV	35,7	124	60,18	2,789	3,418	3,10	—	—	—	[22,23]
	(exp—6)	I	38	123,2	73,7	3,147	3,866	3,443	14,5	14,0	14,7	[25]
	Morse	I	67,1	120	89,7	2,611	3,461	2,908	8,0	5,7	6,9	[26]

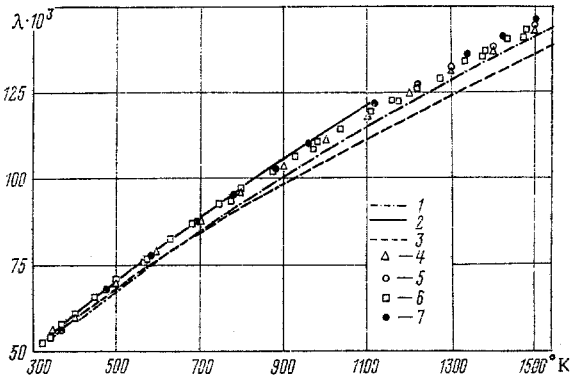

Fig. 1. Temperature dependence of thermal conductivity of neon and comparison of experimental values with theory and data of other authors: 1) calculation (exp-6); 2) data of [7]; 3) [8]; 4) [9]; 5) [10]; 6) [11]; 7) present study. λ, W/m · °K.

The authors of the later studies [9-11] measured the thermal conductivity of neon by the hot wire method with a cold cylinder wall (thermal-diffusion column) in various temperature ranges (350-1500°K [9], 1000-1500°K [10], and 323-2723°K [11]*) to an accuracy of ± 3, 3.2, and 2%, respectively.†

Comparison shows (Fig. 1) that our experimental data agree well (within the limits of experimental error) with the data of [9] over the entire temperature range studied. Our data diverges most from that of [8] (Fig. 1). At T < 900°K the divergence does not exceed 1%, but increases somewhat with increase in temperature (reaching 2% at T = 1500°K). This insignificant increase in our experimental data on neon thermal conductivity can evidently be explained by the presence of impurities in the gas, especially helium (0.1%), while the authors of [9-11] studied spectrally pure gas.

Recommended thermal-conductivity values for neon at atmospheric pressure for the temperature intervals 273-1100 and 273-5000°K, respectively, are presented in [7, 8]. The uncertainity in the data of [7] comprises ± 2% over the entire temperature range, while that of [8] changes with increase in temperature (at T < 500°K, ± 2%, at T > 1000°K, reaching ± 10%).

The recommended neon values of [8] are systematically lower than those of [7] and of the present experiments (Fig. 1). The divergence between the results of [7] and [8] comprises 2.2% $\left(\frac{\lambda_{[7]}-\lambda_{[8]}}{\lambda_{[8]}}\cdot 100\% = 2.2\%\right)$ at T = 400°K and increases with increase in temperature, reaching 13% at T = 1100°K, exceeding the uncertainty of the recommended data themselves.

The experimental values of [11] and our data agree well with the recommended values of [8] at T = 373°K, but with increase in temperature the divergence increases, reaching 7.4% at T = 1503°K.

*Platinum (345-1533°K) and tungsten (323-2723°K) wires were used in the measurement cell in [11].

†The authors of [10] presented only the random and systemic components of the uncertainty. The present authors calculated the total uncertainty of 3.2% by using the recommendations of [12].

TABLE 3. Smoothed and Interpolated Experimental Data on Thermal Conductivity of Neon at Atmospheric Pressure, $\lambda \cdot 10^3$, W/m · °K

T, °K	400	500	600	700	800	900	1000	1100	1200	1300	1400	1500
λ	60,0	69,9	79,2	88,3	96,7	104,8	112,6	119,8	127,0	133,6	140,0	146,0

It is evident from Fig. 1 that all the experimental data agree best with the recommendations of [7]. The greatest deviation (3%) between the values of [7] and the experiments of [9, 10] occurs at 1100°K.

Thus, the recommended values of [7], the experimental values of [9-11] and our own data represent the thermal conductivity of neon over the range 400-1500°K with a satisfactory accuracy. It is preferable to use the data of [7] in calculations up to 1100°K, while for T > 1100°K the values of [9-11] and the present study may be used.

Table 3 presents smoothed and interpolated experimental data on the thermal conductivity of neon in the temperature range 400-1500°K. These are convenient for practical application.

Analysis of theoretical and experimental thermal-conductivity data shows that for calculations in the temperature range 400-1500°K one should not use the Lennard-Jones potential function (12-6) with potential parameters from [23] (giving a divergence of 21-24% over the entire temperature range). The most applicable potential function for calculating the thermal conductivity of neon is the modified Buckingham potential (exp-6) with potential parameters from [24] (greatest divergence between our experimental data and theory does not exceed 4.7%).

At the present time the thermal conductivity of Ne–Ar mixtures has not been studied sufficiently [1] over a wide temperature range, and there are no experimental data for T < 273°K.

The thermal conductivity of an Ne–Ar mixture was first measured in 1954 by the hot-wire method at 273°K to an accuracy of ± 10% [13]. Somewhat later, the authors of [15], using the same method, measured the thermal conductivity of an Ne–Ar mixture at T = 311°K. In [14] experimental values of thermal conductivity of a mixture measured by the hot-wire method (catharometer) at 291°K were presented. The hot-wire method was used in [16] to study thermal conductivity of an Ne–Ar mixture and a number of other binary mixtures of monatomic gases at two temperatures, 302 and 793°K.

In 1967-1968, [17, 18] measured the thermal conductivity of argon–neon mixtures by the hot-wire method to an accuracy of ± 2% in the temperature ranges 313-363°K [17] and 303-363°K [18].

In [19] the thermal conductivity of an Ne–Ar mixture was obtained by a new thermistor method with the quite high accuracy of ± 1% but, unfortunately, at only one temperature, 296.8°K.

The authors of [20] were the first to measure the dependence of thermal conductivity of this mixture on pressure (50-2500 atm) at a temperature of 348°K (accuracy of data obtained, ± 2.5%).

Evidently, together with other difficulties, the traditional appraoch to studying thermal conductivity of mixtures (more accurately, the concentration dependence of thermal conductivity at specified temperatures) has hindered studies over wide temperature ranges. A study of the temperature dependence of thermal conductivity of mixtures of specified composition was initiated in [17, 18] and continued in [4]. The concentration dependence of thermal conductivity was then obtained indirectly from experimental data obtained by studying the temperature dependence of thermal conductivity of mixtures of known composition. This (together with other factors) permitted a significant expansion (to 1500°K) of the temperature range of such studies [4].

In the present study the thermal conductivity of argon–neon mixtures (0.2, 0.4, 0.6, 0.8 Ne) was measured by a molybdenum measurement cell with the following parameters: inner diameter of tube, 5.700 ± 0.005 mm; outer diameter, 6.300 ± 0.003 mm; measurement wire diameter, 0.100 ± 0.001 mm; length of long wire segment, 102.605 ± 0.008 mm; eccentricity, 0.250 ± 0.005 mm; length of short wire segment, 42.909 ± 0.008 mm; resistance of long wire segment at 20°C, 2.5658 ± 0.0004 Ω; resistance of short wire segment at 20°C, 1.0730 ± 0.0002 Ω; effective length 59.696 ± 0.012 mm; effective resistance at 20°C, 1.4928 ± 0.0003 Ω.

The thermal conductivity of the argon–neon mixtures (0.2, 0.4, 0.6, and 0.8 Ne) was measured over the following temperature ranges: 392-1478, 398-1501, 394-1499, and 399-1517°K, respectively.

Figure 2 shows our experimental data for thermal conductivity of neon, argon, and their mixtures, together with the data of [15, 16, 18]. It is evident that our values agree well with the results of [18] at temperatures of the order of 360°K.

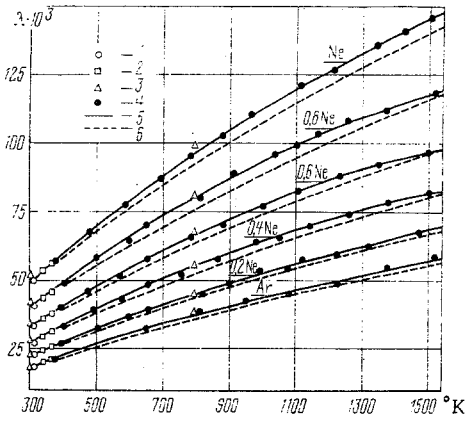

Fig. 2

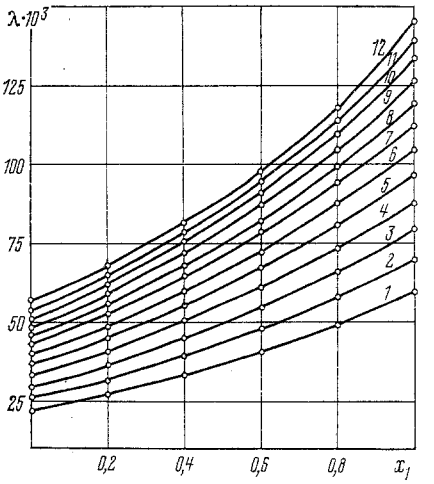

Fig. 3

Fig. 2. Temperature dependence of thermal conductivity of argon–neon mixtures: 1) data of [15]; 2) [18]; 3) [16]; 4) results of present study; 5) smoothed values; 6) calculation (exp-6).

Fig. 3. Concentration dependence of thermal conductivity of Ne–Ar mixture at "round" temperature values: 1) T = 400°K; 2) 500; 3) 600; 4) 700; 5) 800; 6) 900; 7) 1000; 8) 1100; 9) 1200; 10) 1300; 11) 1400; 12) 1500.

Comparison of the present experimental data with the only available values of thermal conductivity at elevated temperature (793°K) for an Ne–Ar mixture [16] shows (Fig. 2) that our data are somewhat low in value. With increase in neon concentration in the mixture the divergence increases. The mean deviation of the four Ne–Ar mixture concentrations of [16] from our data does not exceed 1.5%; i.e., it is within the limits of experimental error of [16].

Table 4 presents graphically smoothed and interpolated experimental values of thermal conductivity of argon–neon mixtures which are convenient for practical use in engineering calculations.

Figure 2 presents theoretical values of thermal conductivity of Ne–Ar mixtures calculated by strict molecular-kinetic theory. Calculations were performed with various intermolecular interaction potentials [Lennard-Jones (12-6), (exp-6), and Morse potential] with various potential parameters (Table 2).

From the comparison of experimental and theoretical values of neon thermal conductivity made above, and a similar comparison for argon [6] in the temperature range 400-1500°K we can conclude that the most applicable potential function for calculation of Ne–Ar mixture thermal conductivity is the (exp-6) potential with potential parameters from [25].

This conclusion is confirmed by the comparison performed between experimental and theoretical thermal conductivity values for Ne–Ar mixtures. The greatest divergence between our experimental data and the values obtained with the (exp-6) potential comprises 5% in the temperature range 900-1100°K. With increase or decrease in temperature the divergence decreases.

The agreement between experiment and calculations obtained with the Lennard-Jones potential (12-6) with potential parameters from [24] and the Morse potential from [26] is not poor.

Figure 3 presents the concentration dependence of thermal conductivity of an Ne–Ar mixture for "round" temperature values. The concentration isotherms deviate negatively from a linear law, which can be explained by the fact that the contribution to heat capacity from the lighter component is reduced due to reduction in the velocity of neon molecules (atoms) upon collision with argon atoms, so that the influence of the heavier component increases.

It is of great interest to establish the effect of temperature (over a wide range) on the amount of deviation from a linear law in the thermal-conductivity-concentration isotherms of Ne–Ar mixtures both from the viewpoint of understanding the physics of transfer phenomena in gas mixtures, and as an aid in solving methodological problems concerning studies of thermal conductivity of binary gas mixtures in engineering practice. We have established experimentally [4, 6] that in the range 400-1500°K the deviation of the concentration isotherms

TABLE 4. Smoothed and Interpolated Values of Thermal Conductivity of Argon–Neon Mixtures at Atmospheric Pressure, $\lambda \cdot 10^3$, W/m · °K

x_i	T, °K											
	400	500	600	700	800	900	1000	1100	1200	1300	1400	1500
0,2	27,4	32,2	36,7	41,0	44,7	48,6	52,2	55,6	58,7	62,1	65,0	68,2
0,4	33,3	39,5	45,0	50,3	55,0	59,8	64,2	68,2	72,0	75,5	78,5	81,4
0,6	40,5	47,8	54,6	60,8	66,6	72,3	77,5	82,2	86,4	90,3	93,7	96,5
0,8	49,0	57,7	65,8	73,1	80,2	87,0	93,4	99,1	104,4	109,0	113,1	117,3

from linearity is independent of temperature, and for an equimolar mixture of He–Ar comprises 40.4%. But this contradicts the experimental data of [16, 21]. Our evaluation of the deviation from linearity for isotherms of an equimolar mixture according to the data of [16] for two temperatures shows that the negative deviation of 40.2% at T = 302°K decreases to 31.9% at T = 793°K, while according to the data of [21] the amount of the deviation increases with increase in temperature.

We will now consider the character of similar deviations for thermal-conductivity values of an Ne–Ar mixture. For the experimental data of [16], with accuracy of ± 2% the deviation of the concentration isotherms from linearity (as in the case of an He–Ar mixture) decreases with growth in temperature from 13.1% at T = 302°K to 10.7% at T = 793°K, while for the data of [17], with an accuracy of ± 1%, with increase in temperature the deviation increases (at T = 313°K the increase is 10.2% and at T = 343°K it is 11.7%). Our experiments on argon–neon thermal conductivity, performed over a wide temperature range, show that the deviation of the concentration isotherms of an equimolar mixture is practically independent of temperature (as for an He–Ar mixture) and comprises about 10.5%.

NOTATION

ΔT_g, true temperature drop in gas layer °K; $\bar{T}$, mean temperature, °K, Q, effective thermal flux, W; Q_t, Q_r, thermal flux transmitted by thermal conductivity and radiation, respectively, W; δT_{sh}, correction for temperature shift, %; λ, thermal conductivity of gas mixture, W/m · °K; x_i, a molar concentration of neon; ε_i, σ_i, ε_{ij}, σ_{ij}, potential function parameters for intermolecular interaction of homogeneous and inhomogeneous molecules; α, slope of exponential repulsion term.

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AN EXPERIMENTAL STUDY OF THERMAL CONDUCTIVITY OF AROMATIC HYDROCARBONS AT HIGH TEMPERATURES AND PRESSURES

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UDC 536.6

Results are presented of an experimental study of thermal conductivity of aromatic hydrocarbons over a wide range of state parameters. Calculated equations which describe the experimental results well are obtained.

There are serious experimental difficulties connected with studying thermal conductivity at high pressures and temperatures. This evidently explains the fact that there is very limited information available in the literature on the thermal conductivity λ of aromatic hydrocarbons at high temperatures and pressures. Data on the values of λ for aromatic hydrocarbons at pressures up to 1500 kg/cm^2 are given only in [1]. Unfortunately even the experimental data of [1] encompass only temperatures below 200°C.

The present study offers results of an experimental investigation of thermal conductivity of m-xylol, n-xylol, o-xylol, and ethylbenzol in the temperature range 30-400°C and pressures to 1000 kg/cm^2. The thermal conductivity of benzol, toluol, and cumol were presented in [2-4].

The characteristics of the hydrocarbons studied are presented in Table 1. The monotonic heating method was employed. The theory of the method, experimental techniques, and device construction were described in [5-7].

The basic component of the apparatus is a cylindrical bicalorimeter, with a gap filled by the liquid to be studied. The inner cylinder (rod) is made of M1 copper. The operating surface of the rod was carefully ground, chrome plated, and polished. The outer cylinder is a massive copper block, in which a tube of 1Kh18N9T is pressed. In contrast to previous construction, a single seal with cone-shaped lip is used to maintain high pressure. The cone angle used is 60°. The bicalorimeter dimensions are as follows: internal diameter of copper block, 11.360 ± 0.005 mm; copper rod diameter, 10.320 ± 0.002 mm, length of bar measurement segment, 100 mm.

Experimental measurement of thermal conductivity reduces to measurement of the time delay of rod temperature relative to temperature of the block. For these measurements a class 0.001 R-345 potentiometer and 51-Sd stopwatch were used, while pressure was generated and measured by an MP-2500 piston manometer, class 0.05, and a set of reference manometers. All corrections essential to the method used [7] were introduced in calculating the thermal conductivity. The maximum calculated uncertainty comprises 2%. Reproducibility of experimental data obtained at one and the same state parameters lies within the limits ± 1%. Possible convection effects were checked by series of experiments at different heating rates (temperature differences). The good agreement of the results indicates the absence of convection. Moreover, for all measurements the product GrPr was significantly less than 1000. Due to the absence of information on absorption spectra the cor-

Ch. Il'drym Azerbaidzhan Polytechnic Institute, Baku. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 33, No. 5, pp. 857-863, November, 1977. Original article submitted December 9, 1976.

 0022-0841/77/3305-1324 $07.50