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STUDY OF OPTICAL PUMPING OF ALKALI ATOMIC BEAM IN STRONG MAGNETIC FIELDS*

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Using rate equation formalism, we got an analytic expression describing the optical pumping of an alkali atomic beam in strong magnetic fields. By comparing with the traditional description in single velocity approximation, the influence of velocity distribution in a practical alkali atomic beam on the pumping effect was quantitatively discussed. It turned out that the rate equation treatment in single velocity approximation might result in an error within twenty percent. Laser optical pumping of ^{7}Li atomic beam in a strong magnetic field (0.15 T) was experimentally studied, and the results are in agreement with the theory in which the velocity distribution is taken into account.

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I. INTRODUCTION

Optical pumping of atoms has been extensively and intensively studied on its significance in physics and its applications. [1-8] For example, it has been frequently used to realize the desired occupation of some special states of atoms and to produce the polarization of atomic beams. [9-8] The theoretical methods so far used to deal with optical pumping process include rate equation [2,3,8] and density operator [9,10] formalisms. When the pumping light has broad line and the light intensity is weak (for a lot of experiments this condition is valid), comparatively simple rate equation formalism is still satisfactory even though the density operator formalism is more general.

Most experiments of optical pumping of alkali atomic beam were conducted in a weak magnetic field, which only served as a quantization axis. In this case, since much more hyperfine Zeeman levels are involved in the pumping process, rate equations have only numerical solutions. Moreover, in earlier treatments it was assumed that atoms in a beam were of uniform velocity, namely, the most probable velocity v_p or average velocity $\bar{v}$, and the influence of atomic velocity distribution (which indeed exists in a practical atomic beam) on pumping process was neglected. This single velocity approximation will bring about some errors.

In the present paper, we investigate, theoretically and experimentally, laser optical

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pumping of alkali atomic beam in strong magnetic fields. In a magnetic field strong enough to decouple the I-J interaction of the atoms under consideration, as it will be seen below, one can get an analytic solution to rate equations, which is a simple function of the pumping time, laser light intensity and polarization. From the analytic solution, the influence of atomic velocity distribution on the pumping effect could be easily taken into account. We first derive the expression of the solution, then give the comparison of experimental results for ^{7}Li pumped in a strong magnetic field with the theoretical ones, and finally discuss the potential applications of the present work.

II. THEORETICAL TREATMENT

Losing no generality, we only deal with the optical pumping process of the alkali atoms with $I=3/2$.

The dependence of the hyperfine sublevels of $S_{1/2}$ ground state and $P_{1/2}$ excited state of the atoms with $I=3/2$ on external magnetic fields is shown in Fig. 1. When

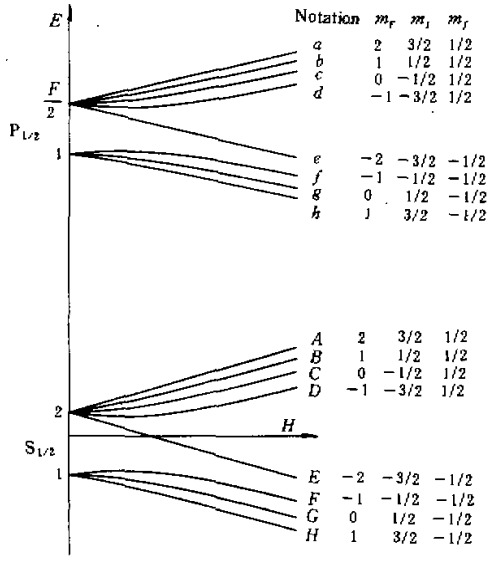

Fig. 1. Hyperfine sublevels of $S_{1/2}$ and $P_{1/2}$ states of alkali atoms with nuclear spin $I=3/2$ (not to scale).

the magnetic field is strong enough, the transitions obey the selection rules: $\triangle m_I=0$, $\triangle m_J=0$ (π transition) and $\triangle m_J=\pm 1$ (σ transitions), and only two ground levels and one excited level are involved in any one of the pumping

processes. For example, in the pumping process $A \rightarrow a$, only A, a and H levels are involved. So we adopt a simple three-energy-level model, as shown in Fig. 2, for this special case. The excited level is labeled by E. One of the two ground levels, labeled by 1 and 2 separately, must be $m_J=+1/2$ state and the other must be $m_J=-1/2$ state; n_1, n_2 and n_e denote the normalized populations of the three levels. C/τ and $(1-C)/\tau$ are the spontaneous transition rates from E to 1 and 2, respectively. For π polarized light pumping $C=1/3$, and for σ polarized light pumping $C=2/3$. Assuming that the level 1 is pumped, then according to rate equations, the time evolutions of n_1, n_2 and n_e satisfy

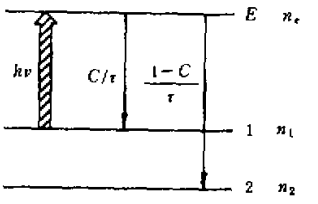

Fig. 2. Three-energy model for dealing with the optical pumping process of alkali atomic beam in a strong magnetic field.

$$\begin{aligned} dn_e/d(t/\tau) &= -n_e + Cx(n_1 - n_e), \\ dn_1/d(t/\tau) &= Cn_e - Cx(n_1 - n_e), \\ dn_2/d(t/\tau) &= (1-C)n_e, \end{aligned} \tag{1}$$

where x is defined as $x=I/2I_0$, I is the pumping light intensity and I_0 the saturation light intensity that can be experimentally determined.[2] By solving Eq. (1) under the initial condition $n_e|_{t=0}=0$ and $n_1|_{t=0}=n_2|_{t=0}=1/N$, where N is the total number of Zeeman levels of $S_{1/2}$ state and $N=8$ when $I=3/2$, the population difference between the two levels 1 and 2, defined as n_2-n_1, after a time duration L/v (L is the width of the laser beam, v is the atomic velocity) of optical pumping can be derived to be

$$\begin{aligned} \triangle n_{sv} = n_2 - n_1 = 2/N - A_1(C,x)\exp[\lambda_1(C,x)L/(v\tau)] \\ - A_2(C,x)\exp[\lambda_2(C,x)L/(v\tau)], \end{aligned} \tag{2}$$

where

$$\lambda_1(C,x) = (1/2)\{-(2Cx+1) + [(2Cx+1)^2 - 4(1-C)Cx]^{1/2}\}, \tag{3}$$

$$\lambda_2(C,x) = (1/2)\{-(2Cx+1) - [(2Cx+1)^2 - 4(1-C)Cx]^{1/2}\}, \tag{4}$$

$$A_1(C,x) = (1/N)\{1 + (2C^2x+1)/[(2Cx+1)^2 - 4(1-C)Cx]^{1/2}\}, \tag{5}$$

$$A_2(C,x)=(1/N)\{1-(2C^2x+1)/[(2Cx+1)^2-4(1-C)Cx]^{1/2}\}. \quad (6)$$

If v in Eq. (2) is replaced by v_p or $\bar{v}$, then Eq. (2) describes the pumping process under single velocity approximation. From Eqs. (3) and (4), we see that $\lambda_1(C,x)<0$ and $\lambda_2(C,x)<0$ always hold, so as the pumping time elapses the 2nd and the 3rd terms on the right side of Eq. (2) will decay, and therefore $\triangle n_{sv}$ increases. If the pumping time is rather long, all the atoms in level 1 will be finally transferred to level 2, corresponding to $\triangle n_{sv}=2/N$.

The atomic velocity distribution in an atomic beam is of the well known form $f(z)=(9/2)z^3\exp[(-3/2)z^2]$ with $z=v/v_p$ ($v_p=(3k_BT/m)^{1/2}$, k_B is Boltzmann constant, T the temperature of the atomic source and m the atom mass). When the influence of atomic velocity distribution is taken into account, obviously the population difference n_2-n_1 should have the following form :

$$\triangle n=n_2-n_1=2/N-A_1(C,x)\int_0^\infty f(z)\exp[\lambda_1(C,x)L/(v_p\tau z)]\,dz$$

$$-A_2(C,x)\int_0^\infty f(z)\exp[\lambda_2(C,x)L/(v_p\tau z)]\,dz \quad (7)$$

III. EXPERIMENTAL

The experimental setup is shown in Fig. 3. The atomic sample used was natural lithium with an isotopic content of 92.58% ^{7}Li and 7.42% ^{6}Li, but only ^{7}Li was under investigation. Lithium atoms effused thermally from the nozzle of an atomic oven with a body temperature of 550 ℃, and were then collimated to form a beam and entered the pumping region in a strong magnetic field of about 0.15 T, which is applied along the direction of the lithium beam, and is strong enough to

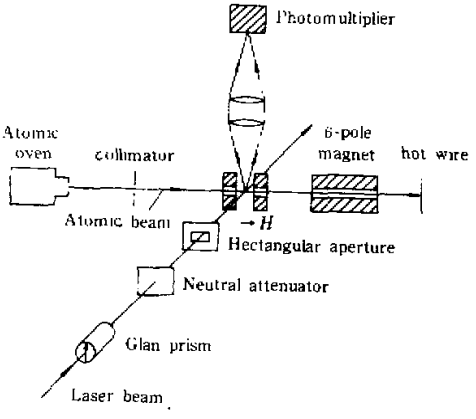

Fig. 3. Experimental setup for optical pumping of ^{7}Li atomic beam in strong magnetic fields.

decouple the *I-J* interaction of lithium. A linearly polarized laser beam (SP 380D ring dye laser) incident upon the pumping region intersected the lithium beam normally with its polarization plane adjustable by Glan prism. By observing the laser-induced fluorescence signal of lithium recorded by a photomultiplier, and adjusting the Glan prism, the desired polarization of the pumping light could be accurately obtained. The width and intensity of the light beam were governed by a rectangular aperture and a neutral attenuator. Optical pumping would lead to atomic population transfer between the two ground levels with $m_J = \pm 1/2$, but the total atomic number or beam intensity remains unchanged. In order to determine the transferred population by means of hot wire surface ionization detector, a hexapole magnet placed between the pumping region and the hot wire was employed[3] for its state-selecting ability to focus the atoms with $m_J = +1/2$ and defocus the atoms with $m_J = -1/2$. When optical pumping results in the transfer of atoms from $m_J = -1/2$ to $m_J = +1/2$ state, the lithium beam intensity transmitted through the magnet will increase; while if the transfer of atoms from $m_J = +1/2$ to $m_J = -1/2$ state takes place, the transmitted lithium beam intensity will decrease. By measuring the transmitted beam intensities with and without optical pumping using hot wire, the population difference (or the atomic polarization[3]) could be determined. After being amplified, the hot wire signal was recorded by a X-Y recorder. The whole atomic beam system was put in a vacuum chamber with the operating vacuum of about 3×10^{-4} Pa.

Two spectra of lithium intensity are shown in Fig. 4, where both σ and π

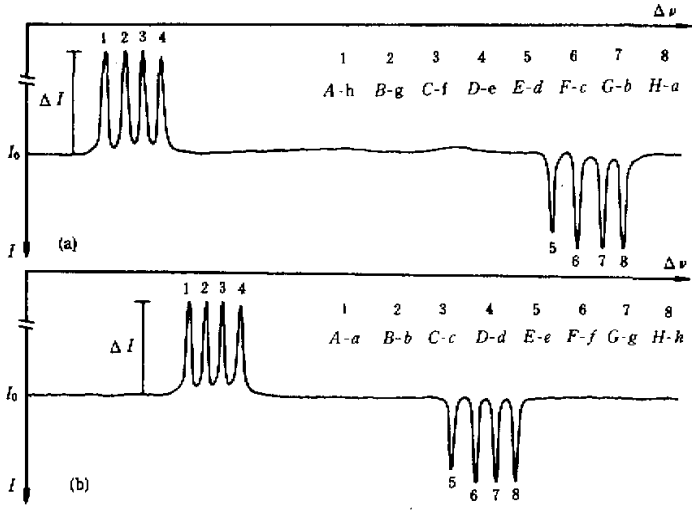

Fig. 4. Lithium beam signals. The insets show the assignments of pumping lines (see Fig. 1). (a) σ pumping, $I = 6.5$ mW/cm^2, $L = 5$ mm; (b) π pumping, $I = 7$ mW/cm^2, $L = 3$ mm.

pumping produce eight transition lines of ^{7}Li with the linewidth of about 35 MHz (^{6}Li signals are very small). As a result of nonlinear scanning of laser frequency, the relative locations of the transition lines were slightly not to scale, but the analyses of the result were not affected. Theoretically, the eight lines should be of the same peak height, but the observed peak heights of the lines 4 and 5 were slightly smaller than the others, which may be due to the residual I-J coupling. Whereas the other six lines had nearly the same peak height, any one of them could be selected for analysis. It has been readily derived that the population difference obeys the relation[11]

$$\triangle n = \triangle I/(I_0 \beta S), \tag{8}$$

where $\triangle I$ is the maximum variation of lithium beam intensity (peak height), I_0 is the lithium intensity without optical pumping, β is the natural abundance of ^{7}Li, and S the selectivity of the hexapole magnet. Assuming that complete transfer of atoms could be achieved when the pumping light intensity is strong and the pumping time is long enough, S could be determined[3, 12] experimentally. In this experiment the measured S is about 0.76.

IV. RESULTS AND DISCUSSION

A. Theoretical results

In Fig. 5 the calculated population differences as a function of pumping time are displayed. In Fig. 5 (a) the pumping time is taken as $L/(v_p\tau)$, while in Fig. 5(b) it is taken as $L/(\bar{v}\tau)$ (in this case v_p in Eq. (7) is replaced by 0.92 $\bar{v}$). The results show that, after a long duration of optical pumping, the complete transfer of atoms from level 1 to level 2 could be achieved; for the same light intensity, π pumping has higher pumping efficiency than σ pumping (by comparing

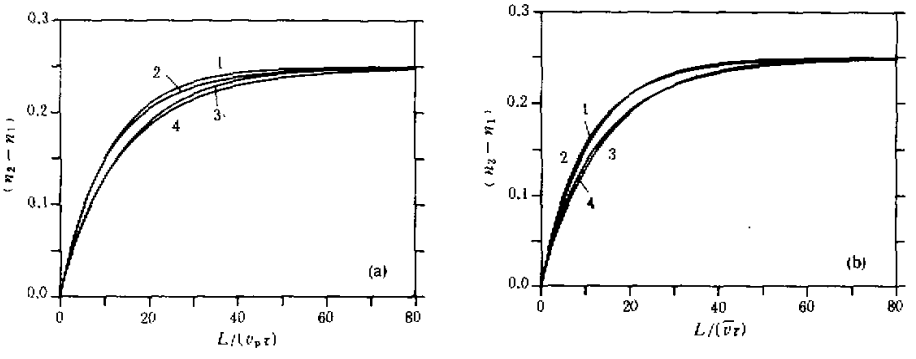

Fig. 5. The dependence of the population difference on pumping time. All the light intensities have been assumed to be equal to the saturation light intensity ($x = 0.5$). The curves 1 and 2 stand for $\triangle n_\pi$ and $\triangle n$ under π pumping respectively; the curves 3 and 4 denote $\triangle n_\sigma$ and $\triangle n$ under σ pumping, respectively.

curve 1 with curve 3, and curve 2 with curve 4); no matter which pumping time is used, the results for single velocity approximation always show deviations from those obtained in consideration of atomic velocity distribution; and for a short pumping time, the deviation is negative, whereas for a long pumping time, the deviation is positive (by comparing curve 1 with curve 2, and curve 3 with curve 4).

To analyze the deviation quantitatively, we give the relative deviation, defined as $(\triangle n_{sv} - \triangle n)/\triangle n$, as a function of pumping time. The results for several different light intensities are shown in Fig. 6. It can be seen from Fig. 6 that, with the increase of the pumping time, the deviation changes from negative to positive

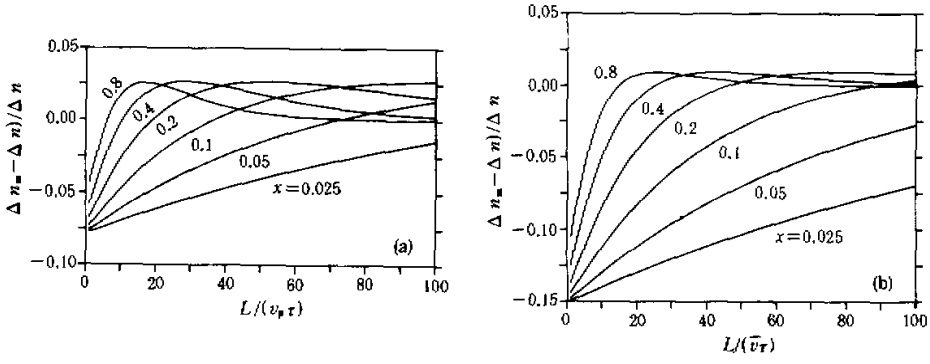

Fig. 6. The deviation of $\triangle n_{sv}$ from $\triangle n$. The pumping light is assumed to be π polarized. The pumping time is (a) $L/(v_p\tau)$, and (b) $L/(\bar{v}\tau)$.

value and finally vanishes. The phenomenon can be explained physically. For a short time of pumping, only slow atoms could be effectively pumped, the pumping time taken in the single velocity approximation is too short for these atoms to be pumped, thus negative deviation results; for a comparatively longer pumping time, because the single velocity approximation assumes a too long pumping time for the fast atoms which are still not effectively pumped, thus a positive deviation is produced. But when the pumping time is extremely long, almost all atoms with different velocities could be effectively pumped, hence the deviation disappears. For the two kinds of pumping times used, $L/(v_p\tau)$ and $L/(\bar{v}\tau)$, the maximum negative deviation (ca. 7%) is smaller in the former case than that in the latter case (ca 15%), and the maximum positive deviation (3%) is greater than that in the latter case (1%). This phenomenon comes from the fact that $\bar{v} > v_p$. From Fig. 6 we also see that the weaker the light intensity, the greater the deviation.

From the above results and discussion, it is clear that the use of single velocity approximation do produce deviation from right description. The deviation ranges from several percent to more than ten percent. Of course, if the pumping process lasts a long time and the pumping light intensity is strong, the deviation

can be neglected. But if the whole process is investigated, the influence of the velocity distribution on pumping effect should be taken into account.

B. Experimental results

The measured and calculated population differences for various light intensities and different light polarizations, are shown in Fig. 7 as a function of pumping time. The points and the solid lines are experimental and theoretical results respectively. Here the pumping time is taken as $L/(v_p\tau)$, and the velocity distribution has been taken into account. From Fig. 7 the following conclusions can be drawn:

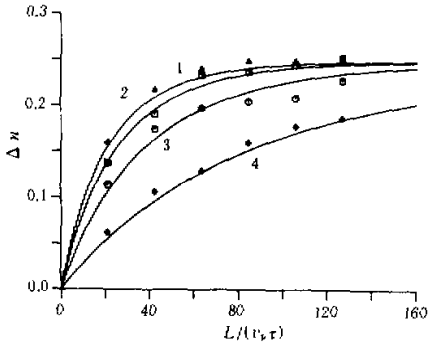

Fig. 7. Measured and calculated population differences as a function of pumping time. 1 (△): π pumping, I=7 mW/cm²; 2 (□): σ, 6.5 mW/cm²; 3 (○): σ, 4 mW/cm²; 4(◇): π, 1.6 mW/cm².

(1). For a fixed pumping light intensity, the pumping efficiency goes higher with the increase of the pumping time; for a fixed pumping time, the increase of the pumping light intensity also helps to raise the pumping efficiency (by comparing curve 1 (△) with 4 (◇), and 2 (□) with 3 (○)).

(2). Under the same experimental condition, π pumping has better pumping efficiency than σ pumping (by comparing curve 1 (△) with 2 (□)).

The above results show clearly that our theoretical treatment for the optical pumping process of alkali atomic beam in a strong magnetic field is reasonable. But it should be pointed out that the deviation introduced by single velocity approximation is small at least in the point of view of experimental measurement. If one wants to measure it accurately, a more accurate detecting means should be used.

Several groups of measured and calculated population differences as a function

of light intensity are shown in Fig. 8. The experimental values are in agreement with the theoretical ones, but no conclusions other than (1) and (2) could be obtained.

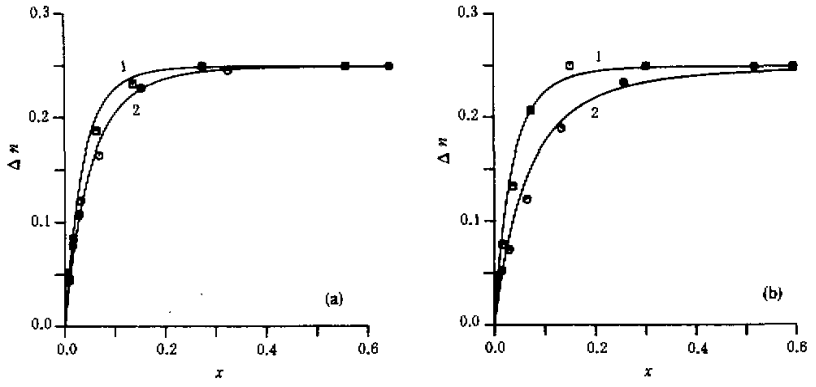

Fig. 8. Measured and calculated population differences as a function of light intensity. (a) π pumping, 1 (⊡) and 2 (⊙) refer to $L=6$ and 4 mm respectively; (b) σ pumping, 1 (⊡) and 2 (⊙) refer to $L=6$ and 3 mm respectively.

V. SUMMARY

In this paper, we have derived an analytic expression describing the optical pumping process of alkali atomic beam in a strong magnetic field directly from solving rate equations. Then based on this expression, the influence of atomic velocity distribution on pumping effect is quantitatively analyzed. It turns out that the conventionally used single velocity approximation may introduce considerable deviations from the correct description. The deviations are explained physically. Finally, the optical pumping of ^{7}Li atomic beam in a strong magnetic field (0.15 T) is experimentally investigated and the results are in agreement with theoretical ones.

The measurements of the repopulations of the ground levels of beam atoms pumped in a strong magnetic field, as shown in this paper, may provide a possibility to determine experimentally the saturation light intensity I_0, and even the lifetime τ, if the measurement is sufficiently accurate. It could be easily verified that the relations $|\lambda_1(C,x)| << |\lambda_2(C,x)|$ and $A_1(C,x) >> A_2(C,x)$ are valid in a wide range of x. This fact means that the dependence of the population difference between levels 1 and 2 on pumping time obeys nearly a single exponential relation. If the population differences for various pumping times and light intensities are accurately measured, then I_0 and τ can be obtained by data fitting. While in a

weak magnetic field, much more levels are involved in pumping processes, the relation between the repopulation of a hyperfine sublevel and the pumping time or light intensity is rather complicated (usually including summation of numerous exponential terms), so it is almost impossible to obtain I_0 and τ by the method described above.

In the present paper the influence of atomic velocity distribution on optical pumping process is demonstrated only for a special case, where the transferred population of atoms has an analytic expression. But as for how to treat the influence on the pumping process of atomic beams in a weak magnetic field, the procedure is more complicated; however, it is still worth investigating.

REFERENCES

[1] W. Happer, *Rev. Mod. Phys.*, **44** (1972), 169.
[2] I. V. Hertel, W. Stoll, *Adv. Atom. Mol. Phys.*, **13** (1977), 113.
[3] D. Hils, W. Jitschin and H. Kleinpoppen, *Appl. Phys.*, **25** (1981), 39.
[4] G. Baum, C. D. Caldwell and W. Schroder. *Appl. Phys.*, **21** (1980), 121.
[5] Zhu Xi-wen, *Acta Phys. Sin.*, **33** (1984), 1605 (in Chinese).
[6] R. N. Watts and C. E. Wieman, *Opt. Commun.*, **57** (1986), 45.
[7] D. Fick, *Appl. Phys.*, **A49** (1989), 343.
[8] P. Cerez and F. Hartmann, *IEEE J. Quant. Electr.*, **QE-13** (1977), 344.
[9] E. de Clercq, M. de Labachellerie, G. Avila, P. Cerez and M. Tetu, *J. Physique*, **45** (1984), 239.
[10] P. Tremblay and C. Jacques, *Phys. Rev.*, **A41** (1990), 4989.
[11] Zhu Xi-wen, Huang Gui-long, Mei Gang-hua and Yang De-lin, *J. Phys. B.* (to be published).
[12] Mei Gang-hua, Zhu Xi-wen and Huang Gui-long, *Acta Phys. Sin.*, **40** (1991), 1776 (in Chinese).