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RESONANCE DIP OF PHOTOIONIZATION IN MAGNESIUM VAPOR

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A new experimental phenomenon - resonance dip(weakening) of multiphoton ionization in Mg vapor with 3s3p ^1P as an intermediate state has been observed. Photoionization and coherent radiation at 5183.7Å, accompanied by ionization dip when Mg vapor pressure was high enough, were studied simultaneously as functions of vapor pressure, laser power density and buffer gas pressure. Possible contributions to ionization dip and coherent radiation are discussed.

It is well known that a resonant intermediatie state will significantly enhance multiphoton ionization [1]. But this is true only when the ratio of vapor pressure to laser power density is very low and only one ionization channel should be considered in Mg vapor. In this note we report a newly observed phenomenon—resonance dip (weakening) of two-photon ionization with 3s3p ^1P as an intermediate state in Mg vapor.

The experiment was carried out by focusing a beam of tunable pulsed UV laser (0.5 – 15mJ/pulse, 4-5ns) produced by a YAG-Dye-SHG of dye laser system (Quanta-Ray DCR2A/PDL-1/WEX-1b) into the center of a 50cm long heat pipe containing Mg vapor and Ar buffer gas. A negatively biased tungsten wire (ϕ=1.5mm) was inserted into the heat pipe to collect the ion signals; the emitted radiation was focused on the entrance slit of a 0.85m double monochromator (SPEX-1404) and was analyzed and detected by a PMT. Both radiation and ionization signals were processed by a two-channel Boxcar integrater (PAR 162/165) and recorded by a chart recorder.

Fig.1 shows the variation of photoionization with the Mg vapor pressure. From fig.1 it can be seen that photoionization was normally enhanced at $3s^2$—$3s3p\ ^1P$ resonance wavelength of 2852Å when Mg vapor pressure was very low ($\leq$0.002 Torr). Keeping laser power density constant and increasing the Mg vapor pressure, a deep dip can be seen near resonance and the wavelength of the dip is 2852Å also. With the increasing of Mg vapor pressure the dip would become wider.

A green fluorescence consisting of three components (5183.7Å, 5172.7Å,

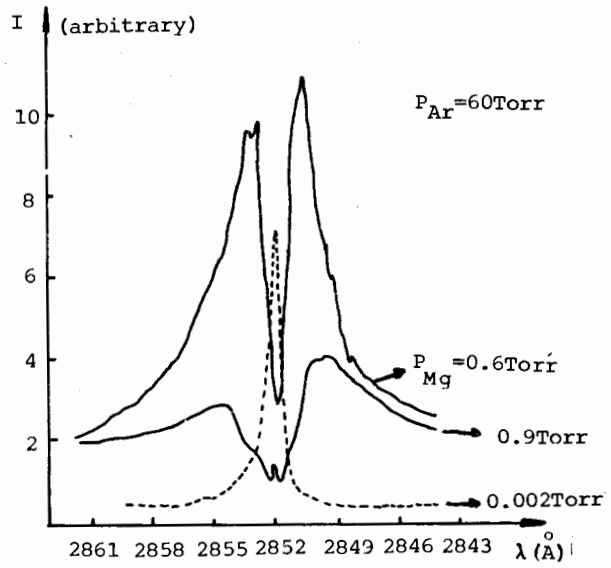

Fig.1 The ion signals as a function of Mg vapor pressure.

5167.4Å) was seen inside the heat pipe as the incident UV laser was tuned over a very wide wavelength range (2770Å to 2960Å). The intensity ratio of individual components agrees with the calculated result[6] of the spontaneous radiation between $3s4s\ ^3S$ and $3s3p\ ^3P$. But when vapor pressure was higher than 0.25Torr, laser power density was higher than $10^8 W/cm^2$ and incident UV laser was tuned near the $3s3p\ ^3P$-$3s5d\ ^3D$ resonance, a highly collimated coherent radiation in the forward direction with a wavelength of 5183.7Å could be observed. It was found that the intensity of the 5183.7Å component was increased by two or three orders of magnitude, but others did not increase very much.

When the Mg vapor pressure was high enough (for example P_{Mg}=1.5Torr), the ion-signal was decreased to very low level. A narrow ionization peak at 2851.6Å would appear when the laser power density was increased. A simultaneous measurement of ionization and coherent radiation at 5183.7Å showed that the ion-peak wavelength corresponds to a dip of coherent radiation as can be seen in fig.2. Further increase of the laser power density at 2851.6Å would make the ion-peak higher and the intensity of the coherent radiation lower.

Another interesting phenomenon is the strong dependence of line shape of ion-signal on the Mg vapor pressure and the laser power density. When Mg vapor pressure was high enough (for example 1.5Torr) the line shape of the broad ion-signal becomes asymmetrical, the ion-signal of the blue side of $3s^2$-$3s3p\ ^1P$ resonance line is much higher than that of the red side. This asymmetry becomes more serious when

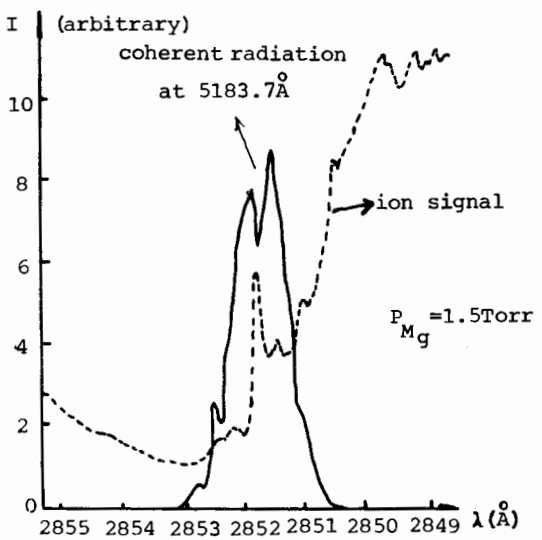

Fig.2 A simultaneous measurement of ionization and coherent radiation at 5183.7Å.

the power density increases. This broad ion-signal and its asymmetry may be related to multiphoton ionization of Mg_2 molecules. Our study on Mg_2 ionization will be discussed elsewhere.

It was observed that the buffer gas pressure obviously affected the ion-signal, the photon absorption and the coherent radiation at 5183.7Å. From fig.3 one can see such an effect on ion-signal.

Resonance dip of laser induced ionization may come from several sources, for instance, transfer of 3s3p ^1P state atoms to other states by collision between two excited 3s3p ^1P atoms. This has been confirmed by fluorescences from some higher states. But the correspondence between the dip of strong coherent radiation at 5183.7Å (which was two or three orders stronger than other fluorescences observed) and the peak of ionization at 2851.6Å (the transition wavelength between 3s3p ^3P and 3s5d ^3D) suggests that the 3s3p ^3P level must be populated and the coherent radiation probably generated by a parametric process, e.g. the incident UV laser photon mixed with stimulated IR photons generated within the Mg vapor by the pump laser itself. The IR radiations could be produced through OPSE or SERS processes by the pump laser.

A direct evidence of the population at 3s3p ^3P was the observation of the transient absorption. Using a pulsed UV pumping laser tuned to the $3s^2$ ^1S—3s3p ^1P resonance and a pulsed background continuum which was well coincided with the pumping laser, a strong transient absorption from 3s3p ^3P to 3s3d ^3D in Mg vapor was observed by Bradley and his colleagues[5].

There are several ways to populate the 3s3p ^3P state when Mg vapor pressure is high enough and laser wavelength is near the $3s^2$-3s3p ^1P resonance. One is the decay from other high states excited by collision, but there would be a delay between input laser and coherent radiation. The experimental results, however, did not show such a delay. The other way is related to the Mg_2 excimer. Inelastic collisions between atoms of excited state 3s3p ^1P and ground state $3s^2$ ^1S can form excimers of Mg_2 ($A^1\Sigma_u^+$ $^1\Pi_g$ $^1\Sigma_g^+$ and $^1\Pi_u$ state)[2,3], predissociation of $A^1\Sigma_u^+$ to repulsive triplet states could occur very fast[4] and then the triplet state molecule could separated into a triplet state 3s3p ^3P atom and a ground state atom (see fig.4). It must be an important way to populate the 3s3p ^3P state via excimer formation and predissociation. If the rate of such a process is larger than that of the two-photon ionization, then the resonance dip would occur.

There is, of course, another channel to ionize the atoms at 3s3p ^3P state by absorbing two photons at 2851.6Å with 3s5d ^3D as a resonance intermediate state. When Mg vapor pressure is high enough, there is a competition between the two-photon ionization with 3s3p ^1P as a resonance

intermediate state and the channel mentioned above. There is also another competition between two-photon ionization via 3s5d ^3D and generation of coherent radiation at 5183.7Å.

We are indebted to Prof.YE Peisuan and Dr.ZHANG Dao-zhong (who observed also the coherent radiation at 5183.7Å in an independent experiment) for helpful discussions and to Dr.K.T.Lu for enlightening discussions and instructions before beginning this work.

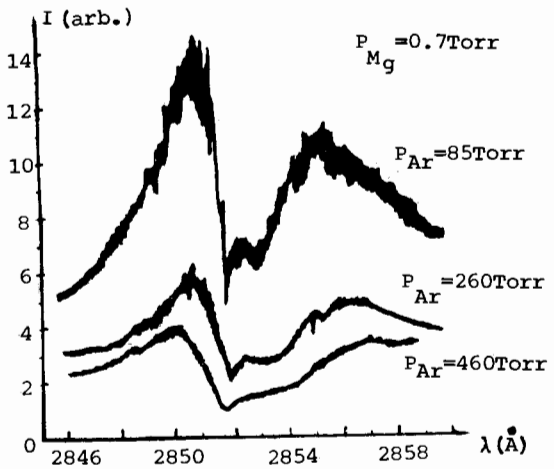

Fig.3 Effect of buffer gas pressure on ionization

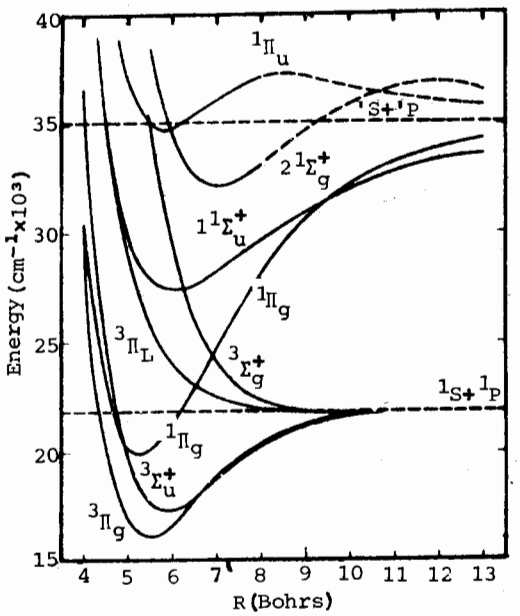

Fig.4 Potential energy curves for the excited valence states of Mg_2

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