APPLICATION OF ION BEAMS FOR SOLID STATE STUDIES

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Abstract

A survey of the works on the non-linear effect in the electronic excitation in ion tracks in solids is given. Following a brief description of the experimental works on the creation of the electron-hole plasma and bi-excitons in CdS and CuCl, the formation of excitons in ion tracks is analyzed. Finally the non-linear effect in anthracene and alkali halides is descussed.

I. Introduction.

Recently, the ion beams have been utilized extensively for the analysis of surface layers of solids.[1] Measurements of the ion-beam backscattering at the surface, of ion-induced X-rays and of nuclear reactions, yield the impurity distribution near the surface. These techniques have been used for studies of corrosion, interfacial reactions, diffusion, etc. Moreover channeling has been shown to be useful for the location of impurities in the lattice.

Ion beams are also known to cause a high-density radiation damage in solids. The studies of radiation damage has attracted much attention because of the recent recognition of the importance of the radiation damage studies for the development of nuclear fusion reactors.[2] The ion-induced surface erosion, sputtering and blistering, is also one of the problems of general interest.

In this review we emphasize another aspect of the usage of ion beams for the solid state studies: the studies of the ion-induced electronic excitation in solids. The main interesting characteristics on the ion-induced electronic excitation is that extremely high density excitation is achieved in ion tracks in solids. Thus several types of non-linear effect have been observed.

II. Creation of the electron-hole plasma in CdS and CuCl

It is known that the excitation of semiconductors by intense lasers produces electron-hole droplets, bi-excitons besides excitons, electrons and holes.[3],[4] Electron-hole droplets are the state where the excitons are generated in such a high density that the excitons form merallic bonding, and the bi-excitons are the molecular state of two excitons. These states of excitons can be idetified because of characteristic luminescence and optical absorption. Extensive works have been accumulated since late 1960's.

Very recently these high density effects have been found to occur in ion tracks in CdS[5] and CuCl.[6] A typical result of the luminescence measurements in CdS is shown in Fig. 1. This result was obtained using low beam current density to avoid the effect of the radiation damage. Two prominent luminescence lines are observed, the relative intensity ratio of which is clearly dependent on the energy and on the kind of particles. Evidently the line at 4870 Å becomes predominant as the stopping power for the particles with higher LET. The line shape and the temperature dependence of these luminescence lines have been studied carefully and shown to be the same as those emitted by the laser excitation.[7] Similarly the bombardment of CuCl with MeV H^+ was found to produce the luminescence lines which are emitted by intense laser excitation. One of these lines at 3918 Å in CuCl had been defenitely assigned to be due to the bi-excitons.[8] Thus it is clear that the bi-excitons are indeed observed in ion tracks in semiconductors.

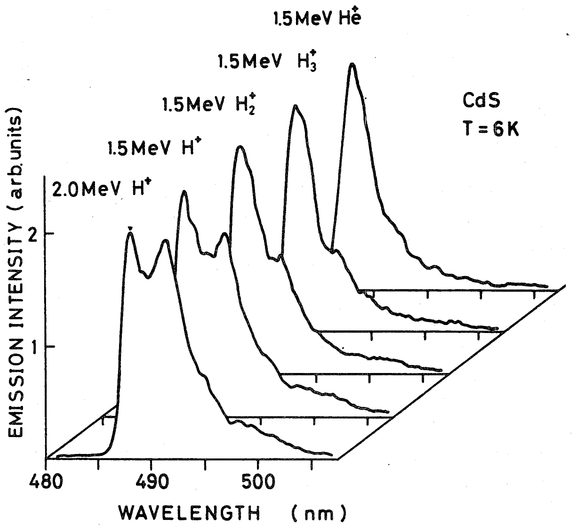

Fig. 1 Luminescence lines induced in ion-tracks in CdS at 10 K.[5]

In spite of a great deal of works on intense laser excitation of CdS and CuCl, luminescence lines due to the electron-hole plasma in CuCl and those due to bi-excitons and electron-hole plasma in CdS have not been identified. One way of the assignment of the luminescence line, which has been used in the earlier studies[7] is to measure the dependence of the intensity of the luminescence line on the intensity of the laser light. At a low light intensity, only the luminescence line due to the isolated excitons are observed. As the intensity of the laser light is increased, they are considered to begin to be replaced by the luminescence line due to the bi-excitons. Further increase in the light intensity could cause the replacement of the bi-exciton line by the electron-hole-plasma line. There are complications upon the above simple arguments: the induced emission and the optical excitation of the bi-excitons or the electron-hole-plasma to the higher states may cause annihilation of the bi-excitons or the electron-hole plasma. In the case of the high-density excitation is ion tracks no such complications arise because of the small volume of the high density region and of the absence of photon field. Thus Mitsushima et al.[5] have assigned that the 4870 Å line in CdS is due to the electron-hole plasma and the other line at 4907 Å due to the bi-excitons. These assignments have been substantiated by the studies of the effect of the applied electric field and of the effect of the lattice defects on the intensity of the luminescence lines.[9]

III. Formation of high density excitation in ion-tracks in solids.

We compare the nature of the high density excitation induced by the laser excitation and that in ion tracks. As pointed out already the volume of the excited region is much smaller in the latter. The other difference is that the spacial distribution of the excited region is homogeneous in the former and inhomogeneous in the latter. We also should note that the energy of excited states is destributed in a δ-function in the former and widely distributed in the latter. The primary energy deposition from the energetic ions will occur within 10 Å around the ion track axis. Since the excitons, bi-excitons and electron-hole plasma are created after they are decerelated to thermal energies, the electrons and holes are considered to be diffused away from the track axis before the excitons are formed. Thus the criterion for the electron-hole plasma and bi-exciton formation in ion tracks is that the exciton concentration after thermalization exceeds the critical concentration, which is approximately 10^{-19} cm^{-3}. It has been shown that the thermalization is completed in 10^{-11} s. On the other hand, for the laser excitation the excitons are accumulated gradually in a specimen. The formation of the electron-hole plasma and bi-excitons will start as soon as the excitons or electron-hole pairs are accumulated above the critical concentration. No high density effect will occur if $I\tau$ is below the critical concentration, where I is the creation rate of excitons per unit volume and τ is the lifetime. It is interesting to note that the creation of the high density excitation in ion tracks occurs adiabatically and that by laser excitation occurs while the equilibrium is maintained.

Since the prominent initial products of the excitation are electron-hole pairs, the delay time in creation of the electron-hole plasma and the bi-excitons involve the thermalization time of the electron-hole pairs and the recombination time of the cluster of electron-hole pairs into excitons. For ion-tracks in CdS and CuCl, the former is much lager than the latter. The spread of the exciton distribution around the track axis occurs mainly due to the kinetic energy imparted upon the excitation. Estimation of the track radius in CdS has been made and shown to be about 200 Å and the concentration have been estimated to be over 10^{19} cm^{-3} for proton tracks. It is also interesting to note that for higher LET ions, the repulsive interaction between excitons may be dominant.

IV. Studies of the high density effect in other solids.

Exciton interaction in ion tracks in organic scintillater has been also studied.[10] It is known taht the particle discrimination can be accomplished using the shape of the scintillation pluse in anthracene. In anthracene the bi-excitons are known to be annihilated due to Auger-type transitions: a bi-exciton formed by two singlet excitons becomes an electron-hole pair, and a bi-exciton fomed by two triplet excitons becomes a singlet exciton. The latter causes a delayed luminescence. Thus the shape of the scintillation pulse is strongly influenced by the local concentration of excitons. The exciton dynamics in the tracks have been analyzed by several authors.[11],[12] These authors put the concentration of excitons in the track after thermalization of electron-hole pairs as parameters. The obtained value for the α-particles is about 10^{19} cm^{-3}. This value is essentially derived as an extrapolation of the delayed fluorescence yield. The concentration after thermalization would be even higher even though detailed calculation has not yet been made.

Studies of the dependence of the scintillation response on LET in NaI(Tl) scintillater has been made extensively.[13] In this case, not only the high density effect but also the yield of the encounter of the excitons with Tl^+ impurities are involved. Recently the effect of LET on the intrinsic luminescence of alkali halides has been made. Kimura and Imamura[14] have found that the ratio of the yield of the singlet luminescence to that of the triplet luminescence is very much dependent on LET. This effect may be related to the mobility, lifetime and the creation yield of the singlet and triplet excitons. It is also of interest to study the effect of the yield of each luminescence on LET. Such work for the triplet excitons has been recently accomplished by Owaki, Matsunami and Itoh and it is found that the yield depends on LET strongly. According to the result of this investigation, reconsideration of the LET dependence of the scintillation in NaI(Tl) is necessary.

V. Concluding remarks.

The arguments made above indicates that the high density excitation in ion tracks produces several types of the non-linear effect. The measurements of the lifetime and the delay time in the luminescence due to bi-excitons and electron-hole plasma may yield important information not only on the solid state physics but also on the track formation in the condense media.

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