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Ionization cross sections of excitons due to scattering by excitons in semiconducting quantum well structures

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Abstract. We have performed theoretical calculations on the ionization cross sections of excitons due to scattering by excitons in semiconducting quantum well structures using a finite confining potential model. It was found that the exciton–exciton ionization cross sections become important for incident energies of the excitons higher than the ionization energy. The magnitude of the ionization cross sections were found to be very much dependent on the type of scattering excitons. Quasi-3D features in the ionization cross sections are also exhibited by excitons in very narrow wells.

1. Introduction

It is well established that excitons play an important role in the optical properties of semiconductors, especially in quantum wells where excitonic features exist up to room temperature. Much work, both experimental and theoretical, has hence been done on determining the exciton linewidths due to various mechanisms, among which exciton–exciton interactions are believed to play major roles in the non-linearities of GaAs quantum wells [1]. Since the linewidth can be related to the cross sections for the interactions concerned, it is hence of interest to calculate the exciton–exciton scattering cross sections.

Elkomoss and Munschy have calculated the elastic [2] and inelastic [3] cross sections due to collisions between identical excitons in bulk semiconductor, for the transitions

$$\mathrm{Exc}(1\mathrm{s}/\mathrm{B}) + \mathrm{Exc}(1\mathrm{s}/\mathrm{A}) \longrightarrow \mathrm{Exc}(1\mathrm{s}/\mathrm{B}) + \mathrm{Exc}(nl/\mathrm{A}) \tag{1}$$

where $nl = 1$s for elastic scattering and 2s, 2p, 3s, 3p, 3d etc, for inelastic scattering and Exc represents exciton. It was found that the inelastic cross sections for transitions with $n = 2$, $l =$s, p, are generally an order of magnitude smaller than the elastic cross sections and are at least two orders of magnitude smaller for transitions where $n = 3$. Similar orders of magnitude have been found for calculations of exciton–exciton scattering cross sections in quantum wells using the exact 2D model [4], together with other interesting features that arise because of quantum confinement. These results give an indication of the relative amount of contribution to the exciton linewidth due to interactions with excitons associated with the various processes described in equation (1).

A similar situation exists for free carrier–exciton scattering [5–7], where it was found that the inelastic scattering cross sections corresponding to the exciton transitions of 1s $\rightarrow$ 2s, 2p, 3s, 3p, 3d are at least an order of magnitude smaller than the elastic scattering cross sections. However, for incident energies higher than the binding energy of the exciton,

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the ionization cross sections for the exciton become important [8–10], as the elastic cross section decreases rapidly for high incident energies. Hence at room temperature, where the thermal energy of the free carriers could exceed the threshold energy for the ionization of the exciton, the interaction of the free carriers with excitons could be dominated by processes whereby excitons are ionized. This is expected to enhance the contribution to the exciton linewidth due to scattering by free carriers at room temperature. Such predictions are consistent for the exact 2D model [8] as well as the quasi-2D models using the infinite [9] and finite [10] confining potential models. Hence it would be relevant to see whether there is an analogue of the above predictions in the case of exciton–exciton interactions.

Recently, we have calculated the elastic cross sections for exciton–exciton scattering using the finite confining potential [11] and showed that excitons in very narrow wells (approximately $0.1a_B$) exhibit a quasi-3D behaviour by the emergence of the trend of bulk exciton–exciton scattering in the elastic cross sections, and that the excitons in the finite quantum wells retain their 2D behaviour for widths larger than about $0.3a_B$, with elastic cross sections similar to those obtained using an exact 2D model [4].

In the present work, we extend the calculation of the exciton–exciton scattering cross sections to include the process in which one of the excitons is ionized, to compare with the elastic interactions. As before, we confine our calculations to a simple two-body problem and use the central field and Born approximations to treat the scattering problem. To simplify calculations, we adopt a semiclassical approach and have neglected effects due to symmetry and exchange of the particles involved. The reasons for these approximations have been addressed in [4], and the keen reader is particularly referred to the series of papers by Elkomoss and Munschy [2, 3, 5, 6]. The effective masses are assumed to be 0.45, 0.085 and 0.068 times the actual electron mass for the heavy hole, light hole and electron, respectively, giving electron-to-hole mass ratios of 0.15 for the heavy-hole exciton and 0.8 for the light-hole exciton. To facilitate comparisons, the units for length are the same as those used in the elastic cross sections, namely, the heavy-hole exciton Bohr radius a_{Bhh} and the light-hole exciton Bohr radius a_{Blh}, and all quantities are expressed in terms of the excitonic units of the ionized exciton.

2. Calculation

The ionization cross sections of excitons due to scattering by free carriers in semiconducting quantum well structures have been discussed in [8] using an exact 2D model and in [9, 10] using a quasi-2D model with an infinite confining potential well model and a finite confining well model, respectively. Here, we use a similar approach to study the ionization of excitons due to scattering by excitons, using the finite confining potential well model, for transitions pertaining to equation (1) where nl now refers to the continuum states.

From previous work on the quasi-2D scattering model, the scattering amplitude can be written as

$$f_{IJ,KL}(\theta) = -\frac{\mu \ \exp(\mathrm{i}\pi/4)}{\sqrt{2\pi k\hbar^2}} \int \mathrm{d}\tau_A \ \mathrm{d}\tau_B \ \mathrm{d}\boldsymbol{R} \ \exp\big[\mathrm{i}\big(\boldsymbol{k}_0 - \boldsymbol{k}\big)\cdot\boldsymbol{R}\big] \ V_i \psi_A^I \psi_B^J (\psi_A^K)^* (\psi_B^L)^* \quad (2)$$

where $\boldsymbol{R}$ is the in-plane relative vector between both excitons and I, J and K, L refer to their initial and final states, respectively. $\boldsymbol{k}_0$ and $\boldsymbol{k}$ are the initial and final relative wavevectors.

We again adopt the variational ground-state wavefunction proposed by Shinozuka and Matsuura [12] which has been found to model closely the behaviour of the exciton for various well widths:

$$\psi_A(\boldsymbol{r}_{a1}, z_1, z_a) = \frac{1}{\sqrt{N_A}} \phi_1(z_1)\phi_a(z_a)\phi_{a1}(\boldsymbol{r}_{a1}, z_1, z_a, \alpha_1, \gamma_1) \tag{3a}$$

$$\psi_B(\boldsymbol{r}_{b2}, z_2, z_b) = \frac{1}{\sqrt{N_B}} \phi_2(z_2)\phi_b(z_b)\phi_{b2}(\boldsymbol{r}_{b2}, z_2, z_b, \alpha_2, \gamma_2) \tag{3b}$$

where 1, 2 refer to the electrons and a, b refer to the holes. $\boldsymbol{r}_{a1}$ and $\boldsymbol{r}_{b2}$ are 2D in-plane vectors between the respective particles. ϕ_i in equations (3a) and (3b) is the envelope function of a charged carrier in a one-dimensional square potential well and is given by

$$\phi_i(z_i) \begin{cases} \cos(k_i z_i) & |z_i| \leqslant L/2 \\ A_i \ \exp(-q_i|z_i|) & |z_i| > L/2 \end{cases} \tag{4}$$

where $i = 1, 2, a, b$ and L is the width of the confining well. According to Shinozuka and Matsuura, an anisotropic hydrogenic wavefunction is used to describe the relative motion between the electron and the hole of an exciton:

$$\phi_{a1}(\boldsymbol{r}_{a1}, z_1, z_a, \alpha_1, \gamma_1) = \exp\left[-\alpha_1\sqrt{r_{a1}^2 + \gamma_1(z_1 - z_a)^2}\right] \tag{5a}$$

$$\phi_{b2}(\boldsymbol{r}_{b2}, z_2, z_b, \alpha_2, \gamma_2) = \exp\left[-\alpha_2\sqrt{r_{b2}^2 + \gamma_2(z_2 - z_b)^2}\right] \tag{5b}$$

where α_1, γ_1, α_2 and γ_2 are variational parameters.

As for the ionized exciton after scattering, we approximated the relative wavefunction of the electron–hole pair by a plane wave in the plane of the quantum well:

$$\psi_{Af}(\boldsymbol{r}_{a1}, z_1, z_a) = \frac{1}{\sqrt{N_{Af}}} \phi_1(z_1)\phi_a(z_a) \ \exp\left(-\mathrm{i}\boldsymbol{k}_{ex} \cdot \boldsymbol{r}_{a1}\right) \tag{6}$$

as discussed in previous work [9]. $\boldsymbol{k}_{ex}$ is the in-plane wavevector between the ionized electron–hole pair which denotes the continuum states.

In the central-field approximation, the interaction potential between the two excitons can be written as

$$V_i = \frac{e^2}{\epsilon}\left(\frac{1}{|\boldsymbol{\rho}_{ab}|} + \frac{1}{|\boldsymbol{\rho}_{12}|} - \frac{1}{|\boldsymbol{\rho}_{a2}|} - \frac{1}{|\boldsymbol{\rho}_{b1}|}\right) \tag{7}$$

where

$$\boldsymbol{\rho}_{ab} = \boldsymbol{R} + s_1\boldsymbol{r}_{a1} - s_2\boldsymbol{r}_{b2} + (z_b - z_a)\hat{\boldsymbol{z}} \tag{8a}$$

$$\boldsymbol{\rho}_{12} = \boldsymbol{R} - s_a\boldsymbol{r}_{a1} + s_b\boldsymbol{r}_{b2} + (z_2 - z_1)\hat{\boldsymbol{z}} \tag{8b}$$

$$\boldsymbol{\rho}_{a2} = \boldsymbol{R} + s_1\boldsymbol{r}_{a1} + s_b\boldsymbol{r}_{b2} + (z_2 - z_a)\hat{\boldsymbol{z}} \tag{8c}$$

$$\boldsymbol{\rho}_{b1} = \boldsymbol{R} - s_a\boldsymbol{r}_{a1} - s_2\boldsymbol{r}_{b2} + (z_b - z_1)\hat{\boldsymbol{z}} \tag{8d}$$

and s_i ($i \in 1, 2, a, b$) is the ratio of the mass of the ith particle to the mass of the exciton to which i belongs, and ϵ is the dielectric constant of the material.

Substituting the wavefunctions given in equations (3a) and (3b) and the interaction potential V_i given in equation (7) into equation (2), the scattering amplitude can be written as the sum of contributions due to interaction between the ith and lth particles as

$$f_{ioniz}(\theta) = \sum_{l,i} f_{li}(\theta) \tag{9}$$

where

$$f_{li}(\theta) = \pm\frac{\mu \ \exp(\mathrm{i}\pi/4)e^2}{\hbar^2\sqrt{2\pi k\epsilon}} \int \mathrm{d}\boldsymbol{R} \ \exp\left[\mathrm{i}\left(\boldsymbol{k}_0 - \boldsymbol{k}\right) \cdot \boldsymbol{R}\right] \left\langle \frac{1}{|\boldsymbol{\rho}_{li}|} \right\rangle \tag{10}$$

and

$$\left\langle \frac{1}{|\boldsymbol{\rho}_{li}|} \right\rangle = \frac{1}{\sqrt{N_A N_{Af}} N_B} \int \mathrm{d}\boldsymbol{r}_{a1}\, \mathrm{d}\boldsymbol{r}_{b2}\, \mathrm{d}z_1\, \mathrm{d}z_2\, \mathrm{d}z_a\, \mathrm{d}z_b\, \phi_1^2(z_1)\phi_2^2(z_2)\phi_a^2(z_a)\phi_b^2(z_b)$$
$$\times \exp\big(\mathrm{i}\boldsymbol{k}_{ex} \cdot \boldsymbol{r}_{a1}\big)\phi_{a1}\big(\boldsymbol{r}_{a1}, z_1, z_a, \alpha_1, \gamma_1\big)\phi_{b2}^2\big(\boldsymbol{r}_{b2}, z_2, z_b, \alpha_2, \gamma_2\big)\frac{1}{|\boldsymbol{\rho}_{li}|} \tag{11}$$

for $l \in a, 1$ and $i \in b, 2$.

The scattering amplitude is a function of the wavevectors $\boldsymbol{k}_0$, $\boldsymbol{k}$ and $\boldsymbol{k}_{ex}$, which are related through the energy conservation constraint

$$k^2 = k_0^2 - \left(\frac{\mu}{\mu_A}\right) k_{ex}^2 - \left(\frac{2\mu}{\hbar^2}\right) E_A \tag{12}$$

where E_A is the binding energy of the ionized exciton, and μ and μ_A are the reduced masses of the combined system and the ionized exciton, respectively. Here we have used the excitonic units of the ionized exciton. For a particular $\boldsymbol{k}_0$ associated with an incident energy higher than E_A, the magnitude of $\boldsymbol{k}_{ex}$ ranges continuously from zero to a maximum given by

$$k_{exmax}^2 = \left(\frac{\mu_A}{\mu}\right) k_0^2 - \left(\frac{2\mu_A}{\hbar^2}\right) E_A. \tag{13}$$

The effective ionization differential cross section is obtained by summing over all the continuum states:

$$\sigma_{ioniz}(\theta) = \frac{k}{k_0} \sum_{\boldsymbol{k}_{ex}} \left| \sum_{l,i} f_{li}(\theta) \right|^2 \tag{14}$$

The total ionization cross section is obtained by integrating all scattering angles numerically:

$$\sigma_{total} = \int_0^{2\pi} \sigma_{ioniz}(\theta)\, \mathrm{d}\theta. \tag{15}$$

3. Results and discussions

Figure 1 shows the total ionization cross sections of the heavy-hole exciton as a function of well width and initial relative wavevector, due to scattering by heavy-hole excitons. Figure 2 shows the corresponding cross sections for the light-hole exciton due to scattering by light-hole excitons. The unit of length used is the exciton Bohr radius of the ionized (target) exciton. In cases where the scattering excitons differ from the ionized exciton, the behaviour and magnitude of the ionization cross sections are similar to those of a similar type of scattering exciton in the identical exciton–exciton scattering and are hence not shown here. The general behaviour of the total ionization cross sections of the exciton due to scattering by excitons is similar to that due to free carriers [8–10] where the total cross sections generally increase steeply after the threshold wavevector to reach a peak, followed by a decrease in the total cross sections as the incident energy further increases. However, the magnitude and characteristics in the trend of the total cross sections due to scattering by excitons seem to be very much dependent on the type of scattering (incident) exciton, in contrast with the case of ionization due to free carriers, where the magnitudes of total cross sections are of about the same order for the different types of incident free carrier. Here, the magnitudes of the total ionization cross sections for the heavy-hole and light-hole excitons due to scattering by heavy-hole excitons range to about $3a_{Bhh}$ and $1a_{Blh}$, respectively, in the larger wells and are roughly two orders larger than those of the respective excitons due to scattering by light-hole excitons, with corresponding magnitudes of about $0.04a_{Bhh}$ and $0.03a_{Blh}$, respectively. Also, when the scattering excitons are light-hole excitons, the total ionization cross sections decrease more rapidly after the peak than for heavy-hole excitons, as can be seen by comparing figures 1 and 2. There is a similar trend in the ionization of excitons by free carriers [9] where the total ionization cross sections decrease rapidly for large k_0 regardless of the type of exciton when the free carrier is an electron or a light hole, while the total ionization cross sections for scattering of heavy holes with heavy-hole or light-hole excitons remain significant for large k_0.

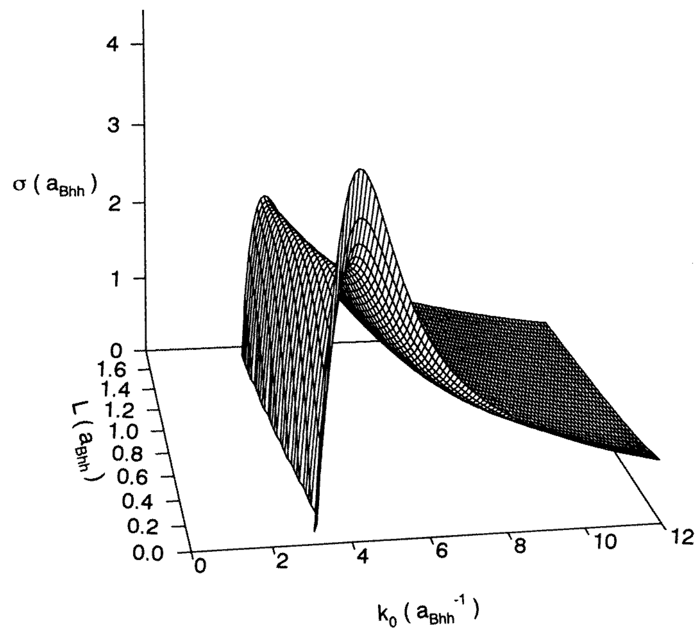

Figure 1. The total ionization cross sections for scattering of heavy-hole excitons from heavy-hole excitons are shown as a function of well width L and initial relative wavevector k_0.

To understand the difference in the orders of magnitude of the total ionization cross sections of excitons due to scattering by light- and heavy-hole excitons, we may first compare the case of ionization of the heavy-hole excitons by heavy-hole excitons and by heavy holes. The ionization cross section of the heavy-hole exciton due to scattering by heavy-hole excitons is smaller than that due to heavy holes by $(2\text{–}3)a_{Bhh}$. The reason for the smaller cross sections due to scattering by excitons than by free carriers is similar to the case of elastic scattering with the scattering amplitude of exciton–exciton interactions being a result of the cancellation of contributions from the composite hole and electron of the scattering excitons with the target excitons, compared with the single electron or hole for free carrier–exciton scattering. Here, the cancellation of the scattering amplitude from the electron of the scattering heavy-hole excitons has roughy halved the ionization cross section of the heavy-hole exciton because of scattering by heavy holes.

We may also compare the present situation with the case of the ionization of hydrogen atoms by incident hydrogen atoms and protons. In [13], the ionization cross sections of the

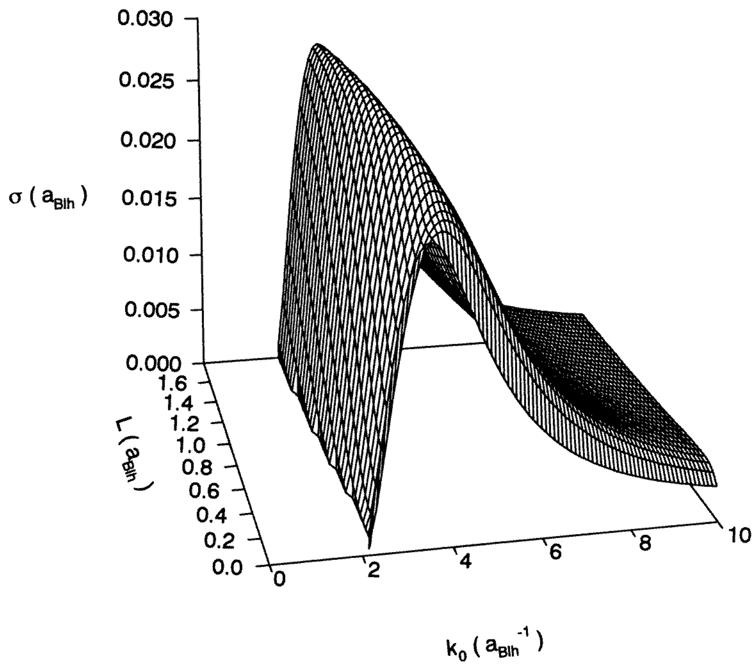

Figure 2. The total ionization cross sections for scattering of light-hole excitons from light-hole excitons are shown as a function of well width L and initial relative wavevector k_0.

hydrogen atoms were investigated for the processes

$$\mathrm{H}^+ + \mathrm{H(1s)} \longrightarrow \mathrm{H}^+ + \mathrm{H}^+ + \mathrm{e}$$

and

$$\mathrm{H(1s)} + \mathrm{H(1s)} \longrightarrow \mathrm{H(1s)} + \mathrm{H}^+ + \mathrm{e}.$$

The results showed that the latter process yields a smaller cross section while retaining similar trends to the former process. Hence our present results of the smaller ionization cross section of the heavy-hole exciton due to scattering by heavy-hole excitons than by heavy holes is reasonable.

Furthermore, if the cancellation of the scattering amplitude due to the composite particles of the incident heavy-hole excitons results in a smaller ionization cross section, then we expect this effect to be more drastic for the case of incident light-hole excitons, as the mass of the light hole is much closer to that of the electron. Hence, the resulting ionization cross sections of excitons due to scattering by light-hole excitons is almost two orders of magnitude smaller than that due to heavy-hole excitons, regardless of the type of exciton being ionized.

To understand the behaviours of the total ionization cross sections of excitons due to scattering by excitons, we may look at the differential cross sections for the various processes. In figures 3 and 4, the differential cross sections for the ionization of the heavy-hole exciton due to scattering by heavy-hole and light-hole excitons, respectively, are shown as functions of the scattering angle θ and initial relative wavevector k_0/a_{Bhh}^{-1} between the excitons, for a well width of $0.5a_{Bhh}$. In figures 5 and 6, the differential ionization cross sections of the light-hole exciton due to scattering by heavy-hole and light-hole excitons,

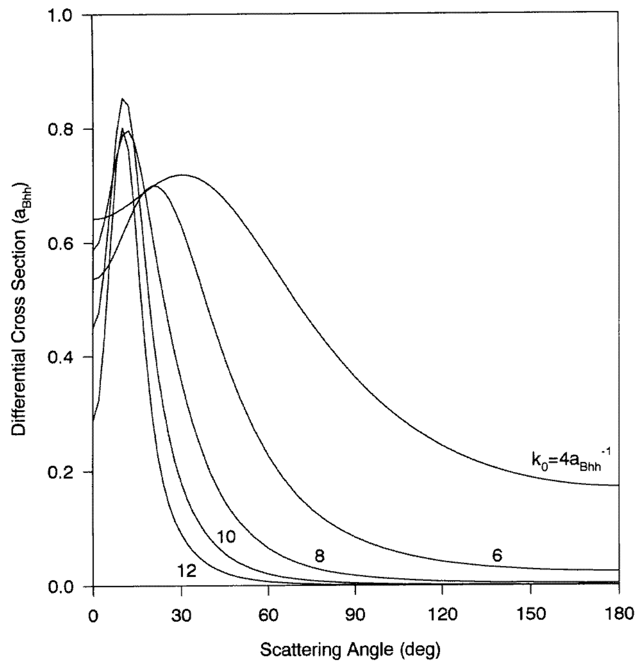

Figure 3. The differential ionization cross section for scattering of heavy-hole excitons from heavy-hole excitons is shown as a function of centre-of-mass scattering angle and the magnitude of the relative wavevector k_0, for a well width of $0.5a_{Bhh}$.

respectively, are shown as a function of the scattering angle θ and initial relative wavevector k_0/a_{Blh}^{-1} between the excitons, for a well width of $0.5a_{Blh}$. We note that the general trends of the differential ionization cross sections of excitons due to scattering by excitons differ significantly from that due to scattering by free carriers shown in [9], even though the trends in their total ionization cross sections are similar. For exciton scattering by free carriers, the differential cross section is generally peaked about the forward direction at $\theta = 0$ for a particular relative wavevector, followed by a monotonic decrease in cross section with increase in θ. However, for exciton scattering by excitons, the differential cross section as a function of θ for a particular incident energy generally starts off at some finite value at $\theta = 0$ and increases to reach a peak, before decreasing with further increase in scattering angle. Also, for larger incident energies, the peak position shifts towards smaller θ with the differential cross section starting at a lower value, while the peak value could either increase or decrease with increasing incident energy, accompanied by strictly decreasing large-angle scattering. The ionization of heavy-hole excitons by heavy-hole excitons exhibits a different trend in the differential cross section with two ‘humps’, compared with the rest of the cases which have only one. There is an analogue of this situation in the ionization cross sections of excitons by free carriers, where the trends in differential cross sections for all types of free carrier are the same except for the heavy holes, which results in ‘kinks’ in the total ionization cross sections. However, for the ionization of heavy-hole excitons by heavy-hole excitons, the peaks in the total ionization cross sections occur at an incident energy (about $4.0a_{Bhh}^{-1}$) corresponding to around the energies where the first hump occurs, while the occurrence of the second hump does not bring about any kinks in the total cross section as the overall decrease in differential cross section with incident energies beyond the second hump slows

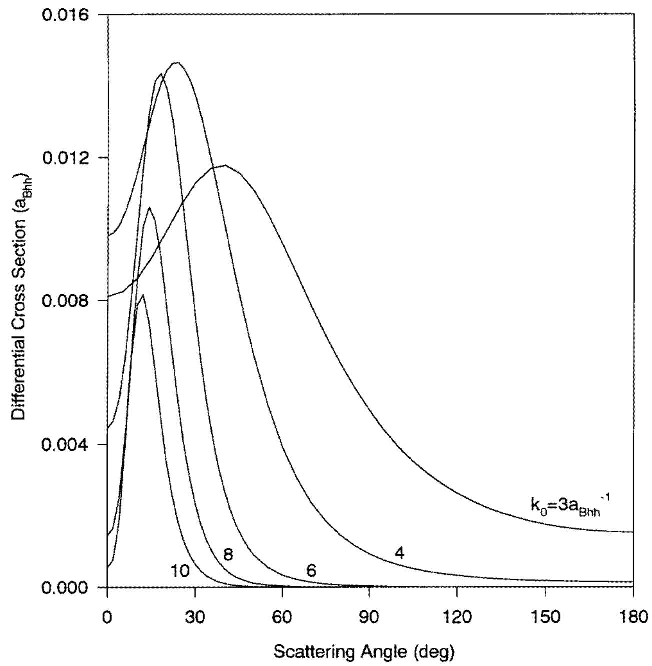

Figure 4. The differential ionization cross section for scattering of heavy-hole excitons from light-hole excitons is shown as a function of centre-of-mass scattering angle and the magnitude of the relative wavevector k_0, for a well width of $0.5a_{Bhh}$.

down to give a gradual decrease in total ionization cross section. For the cases of ionization of light-hole excitons by light- and heavy-hole excitons and the ionization of heavy-hole excitons by light-hole excitons, the peak in the total ionization cross sections occurs at incident energies just before the maxima of the respective humps in their differential cross sections.

The more rapidly decreasing trend in the total ionization cross sections of excitons due to scattering by light-hole excitons may also be seen by comparing the differential cross sections for ionization of the light-hole exciton by heavy- and light-hole excitons (figures 5 and 6). There is generally a faster rate of decrease in differential cross sections with incident energies for all scattering angles due to scattering by light-hole excitons than there is for that by the heavy-hole excitons, which maintain a relatively larger amount of forward scattering even at high energies corresponding to about $10a_{Blh}^{-1}$. This is possibly due to the larger mass of the heavy-hole exciton, and the smaller cancellation effect from the electron of the incident heavy-hole exciton. As for the ionization of the heavy-hole exciton by heavy-hole excitons, although there is a fast decrease in differential cross sections of large scattering angles for energies around the first hump, which hence brought about a rapid decrease just after the peak of the total ionization cross section, it is soon slowed down by the increase in small-angle scattering before the second hump. Again, for the ionization of the heavy-hole excitons, the incident heavy-hole excitons maintained relatively larger forward scattering than did the light-hole excitons. Hence the total ionization cross sections of excitons by heavy-hole excitons remains significant even for large k_0, which is similar to the cases of scattering by heavy holes in the ionization scattering of excitons by free carriers.

As in the elastic exciton–exciton scattering, the peak in the total ionization cross section

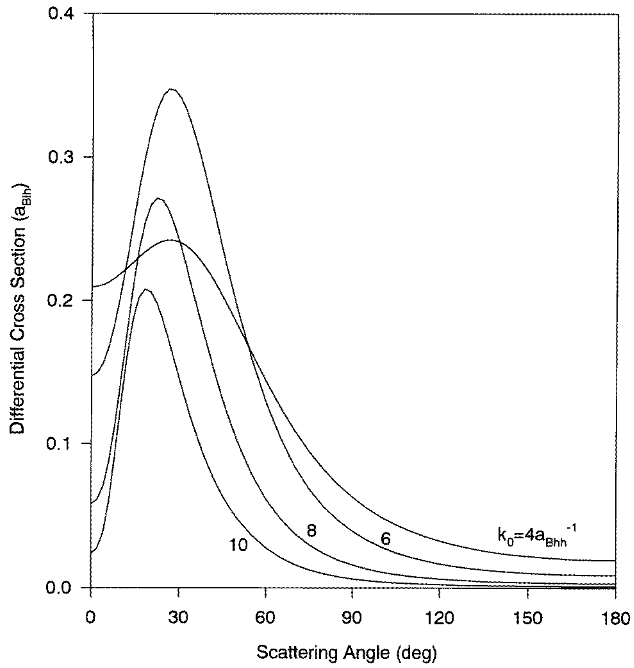

Figure 5. The differential ionization cross section for scattering of light-hole excitons from heavy-hole excitons is shown as a function of centre-of-mass scattering angle and the magnitude of the relative wavevector k_0, for a well width of $0.5a_{Blh}$.

shifts towards smaller incident energies as the well width increases and this is also similar to the case of ionization of excitons by free carriers. Also our previous calculations on exciton scattering by free carriers and excitons show that there are generally enhancements in cross sections below a certain thin well limit with the emerging trend of the corresponding bulk exciton scattering cross sections. Here, the excitons in the very narrow wells again show enhancement in the total ionization cross sections with the cases of incident heavy-hole excitons showing obvious increase in total cross sections while the cases of incident light-hole excitons showed increased cross sections only for the smaller incident energies but, nevertheless, with the corresponding shift in the peak of the total cross sections towards smaller incident energies, which reflects an emerging 3D behaviour of the excitons. As discussed in previous work, such features are associated with the quasi-3D behaviour of the excitons in very narrow wells (about 10 Å) where the total energy of the exciton becomes comparable with the confinement potential which hence allows substantial smearing of the exciton wavefunction into the barrier regions. The 2D exciton under confinement then begins to gain dimensionality along the direction of confinement, exhibiting a quasi-3D behaviour.

Quasi-3D excitons in very narrow wells have been proposed by several workers in their calculations of the binding energy of excitons under confinement of a finite potential well when there is a decrease in binding energy below the quasi-3D limit. Here, together with our previous studies, we again show that such quasi-3D behaviour can also be deduced from the cross sections which indirectly reflect the nature of the excitons.

In comparison with previous calculations on exciton–exciton elastic scattering and exciton–free carrier scattering, the total ionization cross sections are generally of the same

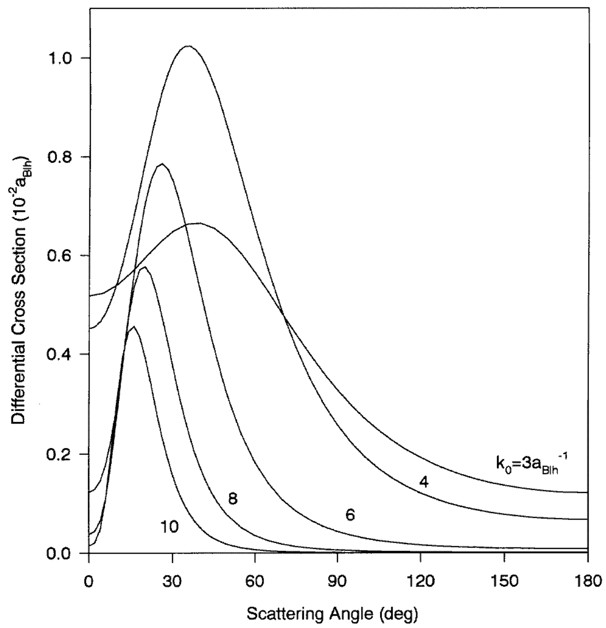

Figure 6. The differential ionization cross section for scattering of light-hole excitons from light-hole excitons is shown as a function of centre-of-mass scattering angle and the magnitude of the relative wavevector k_0, for a well width of $0.5a_{Blh}$.

order of magnitude as the corresponding elastic cross sections. However, the ionization cross sections are associated with higher incident energies where the elastic cross sections, which decrease rapidly with increasing incident energy, are small. Hence, in situations where the thermal energies of the excitons exceed the threshold energy for ionization to take place, the exciton–exciton interaction would be dominated by processes which result in the ionization of the excitons into electron–hole pairs. This again should contribute significantly to the portion of the exciton linewidth due to interactions by excitons. In contrast, at lower temperatures, exciton–exciton interactions would be dominated by elastic scattering, as in the free carrier–exciton scattering.

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