Surface Science 226 (1990) 191–198
North-Holland

CARRIER EXCITATION BY ATOMIC COLLISION ON GaAs(110)

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Received 9 August 1989; accepted for publication 20 September 1989

We have measured the excitation yield for carriers produced at the outermost atomic layer of a GaAs(110) surface by impinging hyperthermal Xe and Kr neutral atoms. The excitation yield roughly scales as exponential in the inverse of the mean energy deposited in the collisions. This is consistent with previous results for InP, which indicated a rapid electronic equilibration with the local lattice kinetic energy dissipated in the vicinity of the atomic impact. However, for GaAs(110) the maximum in the excitation yield, as a function of incident angle, is not normal to the surface. Instead, the maximum yield is found for polar angles nearly perpendicular to the tilted Ga–As surface atom bonds. In addition the yield is two orders of magnitude greater than that estimated from equilibrium arguments. These results are in apparent disagreement with the simplest ideas of local equilibration, and may provide the first insights into the collisional excitation mechanism of electrons by neutral atoms.

1. Introduction

In the exchange of energy between an atom or a molecule and a solid surface, it has long been of interest to understand what fraction directly involves the electronic degrees of freedom of the solid [1]. Recently we have carried out experiments in which we directly measure the probability of electronic excitation in atom–surface collisions and the corresponding fraction of the total energy transfer which goes into the electronic degrees of freedom [2–5]. Model systems were chosen for study – hyperthermal rare gas atoms incident upon high resistivity semiconductor surfaces. We have previously studied the collisional excitation of electron–hole pairs (e^-h^+) by Xe and Kr on InP(100) and InP(110) [3–5]. Prior to that, we had observed collisional excitation of e^-h^+ for Xe on Ge(100) [2]. In this paper we report the excitation of e^-h^+ on the GaAs(110) surface.

GaAs(110) has certain advantages as a target. It has a well-established geometric and electronic structure [6–9], and the surface can be maintained in a well-ordered and stoichiometric state with standard surface processing techniques. In addition, we have previously carried out scattering experiments for rare gas atoms over the same incident energy range on GaAs(110) [10], and complementary trajectory calculations of the scattering have also been performed [11]. Thus the GaAs(110) surface offers the prospect of a detailed understanding of the excitation mechanism as the physical properties of the surface and the scattering dynamics are well-established. In contrast, a disadvantage arises from the shorter carrier recombination lifetimes for GaAs compared to InP which lead to significantly smaller signals.

The experiments described here provide a test of the simple ideas (reviewed below) which seemed to adequately describe our previous e^-h^+ excitation data for InP(100) and InP(110) [3,5]. Specifically, we postulated a rapid transient equilibration between the energy transferred to the local atomic environment immediately surrounding the impact point and the electronic degrees of freedom of

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that region. This was based on the finding in refs. [3,5] that the energy dependence of the collisional excitation yield (Y) for Xe and Kr on the InP(100) was well described by a Boltzmann form, i.e.

$$Y = A \exp\left(-\frac{3nE_{bg}}{\overline{\Delta E}_{RG}}\right), \quad (1)$$

where A is a constant, n is the (experimentally derived) effective number of atoms involved in the electronic equilibration, E_{bg} is the band gap of the semiconductor, and $\overline{\Delta E}_{RG}$ is the mean energy deposited by the rare gas atoms in the collision. This expression is simply a Boltzmann ratio for the occupation above the bandgap in a local region of n atoms excited to a local temperature $T_\ell \sim \Delta E/C_p$.

We suggested two approximations for the preexponential A based either on intrinsic carrier excitation at the effective local lattice temperature (T_ℓ) or on atomic ionization at fixed volume. The yield estimated either way was found to be in good agreement with the excitation yield measured for InP (assuming that the effective number of atoms derived for InP(100) ($n = 8$) was also appropriate for InP(110)) [5]. For both of these surfaces the carrier lifetimes were found to be determined by the surface recombination rates. This allowed us to use the carrier lifetimes, which we determined for these samples by optical excitation, for the carriers excited only at the surface by atomic impact, and thereby obtain the absolute values of the collisional excitation yield.

In this paper we find that the yield of e^-h^+ for the GaAs(110) surface due to hyperthermal atomic impact is two orders of magnitude higher than that expected if the simple equilibration model were to hold. We further find the probability of excitation to be highest for collisions in which the surface Ga atom is struck directly, rather than maximizing at the surface normal, as we had found previously for InP surfaces. We suggest that these results are considerably different than what is expected for an equilibrated region. They are to some extent characteristic of the dynamics of the excitation process and may motivate further (theoretical) interest into the excitation mechanism.

2. Experimental

The experiments were conducted in an ultrahigh vacuum (UHV) chamber described previously [5,12]. Atomic beams of Xe or Kr seeded in H_2 were expanded through a heated nozzle. The beam energies could be varied from 2–10 eV by varying the rare gas/hydrogen ratio and the nozzle temperature. The speed ratios for these beams ranged from 13–20 ($= v/\Delta v$ full width at half maximum).

The samples used in the experiments were defect-compensated semi-insulating GaAs(110) single crystals. The GaAs resistivities were typically 1×10^6–2×10^8 Ω cm. The crystals were prepared by sputtering with 500 eV Ar^+ at room temperature and then annealing to ~ 600 °C. Helium diffraction spectra were observed from these crystals, and compared favorably to previously published data for this surface [8].

Conductance measurements were performed as described in ref. [3] by applying a bias voltage across ohmic contacts on the front surface of the crystal. The modulated molecular beam illuminated a small area in between the two contacts, and the modulated transient current due to the incident atoms was recorded by a lock-in amplifier. The small modulated photoconductance, due to the blackbody radiation from the heated molecular beam nozzle, was measured by recording the modulated currents when a neat H_2 beam was incident on the crystal at the same nozzle temperatures as for the rare gas/H_2 mixtures. The photoconductive contributions were then subtracted from the signal observed for the rare gas/H_2 beam experiment to give the collisional excitation signal. The GaAs(110) single crystals used were 0.023 cm thick, the edge of the contacts were separated by the target area which was 0.3 cm long and 0.5 cm wide. The crystals were supported on a sapphire block to provide electrical isolation from the crystal manipulator.

As in refs. [3,5] the excitation yield (Y) is defined to be the collision generation rate of carriers (G) per incident flux of rare gas atoms (F_{RG}):

$$Y = G/F_{RG}. \quad (2)$$

It was assumed that during each rare gas atomic beam pulse the excitation current quickly reached steady state, so that the sample-averaged collisionally-generated excess carrier density is given by [#1]:

$$\Delta N = G\tau/V, \tag{3}$$

where τ is an effective carrier recombination lifetime and V is the volume of the crystal over which the conductance is measured (the volume between the contacts). As described below these lifetimes can be estimated by optically exciting carriers under conditions similar to the collisional excitation. The excess carrier density is measured by the change in conductance of the crystal which is related by a geometrical factor to the change in conductivity ($\Delta\sigma$):

$$\Delta\sigma = q\mu\ \Delta N, \tag{4}$$

where μ is the sum of the electron and hole mobilities for GaAs. We have used $\mu_n = 8000$ $cm^2/V \cdot s$ and $\mu_p = 320$ $cm^2/V \cdot s$ for the electron and hole mobilities of GaAs, respectively [14]. The yield is then given by [#1]:

$$Y = \Delta\sigma V/q\mu\tau F_{RG}. \tag{5}$$

Carrier recombination lifetimes were measured following the procedure described in ref. [5]. Carriers were optically excited using a HeNe laser at 632 nm, or a HeCd laser at 442 nm. In both cases, the corresponding photon energies are greater than the direct band gap of GaAs, $E_{bg} = 1.42$ eV [14]. The incident laser powers were varied from 1 nW to 10 mW using a series of neutral density filters. The spot size on the crystal was 0.1 cm. Typically, the laser was opened to the crystal for a few milliseconds at a repetition rate of less than 1 Hz. A voltage bias was applied across the contacts and the photoconductivity signal was amplified and then recorded on a multichannel signal averager in summation averaging mode. The measurements were repeated 100 times at each laser power. The steady state difference in the number of carriers and the generation rate due to the known laser power absorbed were used to determine the effective minority carrier lifetime as a function of the sample-averaged carrier density [5,15]. The data were fit to a curve of the form suggested by Landsberg [16]:

$$1/\tau = 1/\tau_{SRH} + a_1 n + a_2 n^2, \tag{6}$$

where τ is the bulk-averaged carrier recombination lifetime measured, n is the excess carrier concentration due to optical excitation, and a_1, a_2, and τ_{SRH} are (non-negative) constants fit to the data. τ_{SRH} is known as the Shockley–Read–Hall lifetime, appropriate for low carrier densities, and is discussed later.

The signal to noise in both the collisional and optical excitation experiments were found to be limited by persistent low frequency oscillations of the type described for semi-insulating GaAs in ref. [17]. We found that the oscillations were significantly enhanced when light was incident upon the crystal. As in ref. [17] photoconductivity was found to be peaked near the contacts. For this reason, the optical and collisional experiments were performed at the center of the region between the contacts.

We have checked the assumption that the collisional carrier excitation reached steady state during the measurement by recording the modulated conductance at modulation frequencies from 80 to 500 Hz. No frequency dependence was found. In addition, the time dependence of the collisional excitation signal was recorded on a multichannel signal averager and coincided with the time of arrival distribution of the rare gas atoms at the crystal surface.

3. Results and discussion

3.1. Incident energy dependence of excitation yield

The excitation yield per incident atom is shown for Xe and Kr as a function of incident energy (E_i) for normal incidence ($\theta_i = 0°$) in fig. 1. The relative yield is defined as the transient current divided by the incident flux of rare gas atoms. The excitation yield rises with increasing incident energy, and is higher for Xe than for Kr at each incident energy.

[#1] Note that eqs. (7) and (9) of ref. [5] should read as eqs. (3) and (5) here, respectively. Similarly, eqs. (2) and (4) of ref. [13] are also missing V, the effective sample volume.

In ref. [5] the mean energy transfer in the atom–surface collision, $\overline{\Delta E}_{RG}$, of (1) was assumed to be $\overline{\Delta E}_{RG} \approx k_{RG} E_i \cos^2\theta_i$. This assumption was based upon the results of ref. [10] in which the mean energy transfer was found to be well fit by the expression $\overline{\Delta E}_{RG} \approx k_{RG} E_i \cos^2[(\theta_i + \theta_r)/2]$. It was further found that $k_{Xe} > k_{Kr} > k_{Ar}$. The correlation found in ref. [10] suggests that the mean energy transfer is proportional to the energy associated with motion normal to the surface, and further that the scattering is approximately locally specular. Using this assumed mean energy transfer, in fig. 2 we show the data of fig. 1, plotted as $\log(Y)$ versus the inverse of the mean energy transfer, is higher for Xe than for Kr at each incident energy, so that the two sets of data are moved on to (or close to) one line on this plot (as in ref. [5], values of $k_{Xe} = 1.00$ and $k_{Kr} = 0.90$ were taken [4]). The data can be fit approximately by a straight line, however, the Kr data appear to be systematically lower than the expected from the Xe data.

These data are approximately consistent in form with what is expected if the yield is determined by a rapid electronic equilibration with the local lattice kinetic energy. The least squares fit of a straight line to the data of fig. 2 gives a slope of -27 eV which can be used to determine the effective number of atoms involved in the electron equilibrium (n). Using eq. (1), and the band gap of GaAs, $E_{bg} = 1.42$ eV [14], we obtain $n = (-27\ \text{eV})/[-3(1.42\ \text{eV})] \approx 6$ atoms. In comparison, for InP we obtained a slope of -32 eV which gives a similar value of $n \approx 8$ atoms [5].

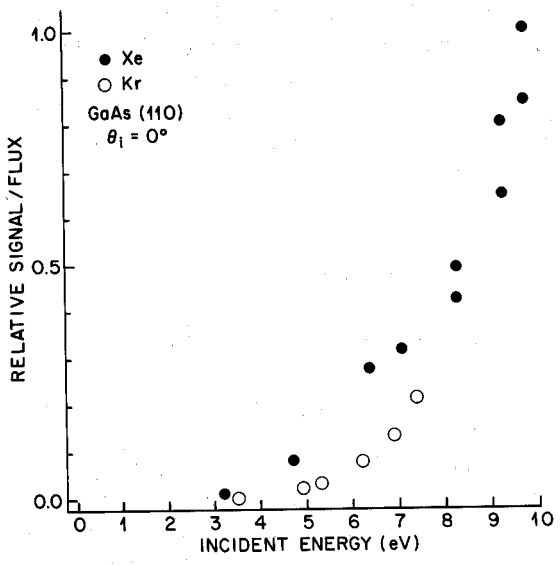

Fig. 1. Electron–hole pair excitation yield per incident atom as a function of incident energy for Xe and Kr at normal incidence on GaAs(110).

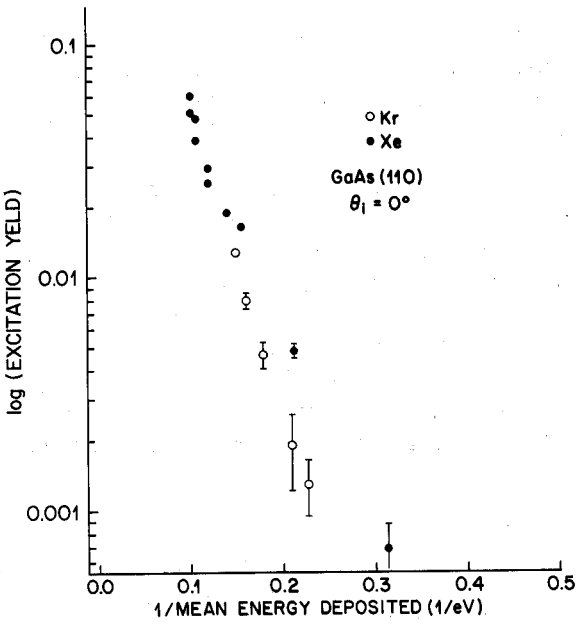

Fig. 2. Relative electron–hole pair excitation yield per incident atom as a function of the inverse of the energy deposited in the collision for Xe and Kr at normal incidence on GaAs(110).

3.2. Absolute excitation yield

The absolute yield was derived in ref. [5] using carrier recombination rates derived from optical carrier excitation as discussed above. The effective carrier recombination lifetimes were measured over a range of excess carrier densities extending down to those found in the collisional excitation experiments. A typical measurement of effective carrier recombination lifetime versus the sample-averaged excess carrier density is shown in fig. 3. In all cases the carrier lifetimes were found to be dominated by the surface recombination rates. We argue that this allows us to make use of the lifetimes of these carriers, optically excited

throughout the laser penetration depth, for the carriers excited only at the surface by atomic impact. From eq. (5), the absolute yield for Xe at $E_i = 9.3$ eV and $\theta_i = 0°$ was found to be $Y = 0.06$ e^-h^+/Xe. Note that this yield means that slightly more than 1% of the total energy transfer is attributable to direct electronic excitation. However, for the 6% of the collisions for which electronic excitation occurs, > 15% of the energy transferred is in direct electronic excitation.

The uncertainty in this value of Y is dominated by the uncertainty in our measure of the carrier recombination lifetime via optical excitation, which is probably good to within a factor of two. Differences in beam shape – rectangular with a flat distribution for the atomic beam, and circular with a Gaussian (TEM_{00}) distribution for the laser beam – made it difficult to have identical spot sizes for the most directly comparable excitation yields.

By estimating the pre-exponential of eq. (1), the idea of a rapid electronic equilibration with the local lattice kinetic energy can be further tested. As proposed in ref. [5], we consider two analogies for determining the pre-exponential A. The first is based on intrinsic carrier excitation:

$$A = 2\left(\frac{2\pi kT_\ell \sqrt{m_e^* m_h^*}}{h^2}\right)^{3/2} V_n, \qquad (7)$$

where m_e^* and m_h^* are the effective masses of electrons and holes, respectively, for the semiconductor, V_n is the volume of the n atoms and T_ℓ is the local lattice temperature determined by the energy transfer. This second is appropriate for atomic ionization at fixed density:

$$A = 2\frac{g_i^+}{g_k}\left[\frac{2\pi kT_\ell}{h^2}\left(\frac{m_e^* m_h^*}{m_e^* + m_h^*}\right)\right]^{3/2} V_n, \qquad (8)$$

where g_i^+ and g_k are the degeneracies of the neutral and ionized states, respectively, and are assumed to be equal [#2]. Assuming that for the highest incident energy conditions, $E_i = 9.8$ eV for Xe at normal incidence, $\overline{\Delta E}_{Xe} = 8$ eV, we find that $T_\ell = T_0 + \overline{\Delta E}/3nk = 5200$ K. Using the density of GaAs, $\rho = 5.317$ g/cm^3 [14], $V_{n=6} = 1.35 \times 10^{-22}$ cm^3, and the effective masses of electrons and holes in GaAs, eqs. (2) and (3) give pre-exponentials of $A = 1.8 \times 10^{-2}$ and 3.2×10^{-3}, respectively, and corresponding yields of $Y = 8 \times 10^{-4}$ and 1.3×10^{-4}, respectively. These values are significantly lower than the experimentally derived value of $Y = 0.06$ e^-h^+/Xe.

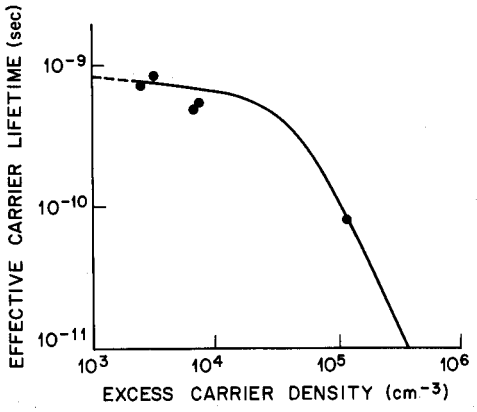

Fig. 3. Effective minority carrier lifetime versus sample-averaged excess carrier density for GaAs(110).

Note that in the plot of effective carrier recombination lifetime versus excess carrier density shown in fig. 3, the measured effective carrier recombination lifetime continues to rise with decreasing carrier density at the low excess carrier densities measured in the collisional experiments. This is in contrast to InP, where it was found that the constant Shockley–Read–Hall lifetime, τ_{SRH}, dominated at low carrier densities and thus throughout the range of the collisional excitation experiments. At the calibration point of $E_i = 9.8$ eV and $\theta_i = 0°$, the lifetimes used (fig. 2) were those measured at the same carrier density. If the carrier lifetimes continued to rise at lower densities, then the actual yield versus energy deposited would fall off faster than shown in the figure. This would have the effect of lowering the estimated yields from the equilibrium argument further eqs. (1)–(3), and a greater deviation from the experi-

[#2] In ref. [5], the electron–hole pair reduced mass was taken as the effective mass of the electron because of the large discrepancy between m_e^* and m_h^* for InP. Here m_e^* ($= 0.063$ m_e) and m_h^* ($= 0.50$ m_e) are similar enough that m_h^* cannot be neglected.

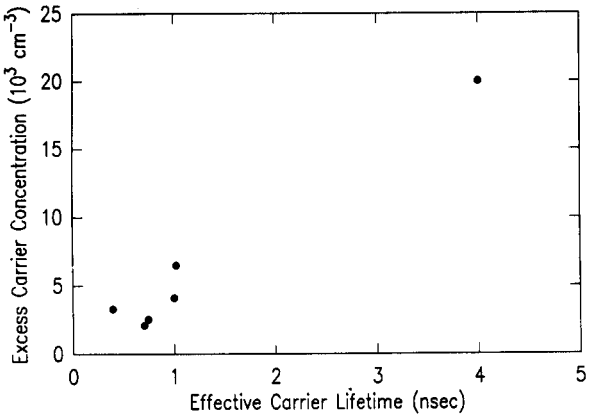

Fig. 4. Collisionally excited excess carrier density (sample-averaged) versus effective minority carrier lifetime for Xe on GaAs(110) at 9.8 eV and normal incidence.

mental values found because of the dependence of the pre-exponentials on the effective number of atoms involved in a local electronic equilibration (from eqs. (7), (8), $A \propto T^{3/2} V_n \propto n^{-3/2}$).

The measured carrier lifetimes were in the range $0.4 < \tau < 4.0$ ns and appeared to depend upon the quality of the surface preparation. Note that at the high end of this range the measured lifetimes were essentially the bulk lifetime for holes (the minority carrier here) in GaAs [18], implying that the surface contribution to recombination was small where these lifetimes were recorded.

In ref. [5] it was assumed that the optically-derived lifetime was appropriate for the determination of the absolute excitation yield using eq. (5). That is, it was assumed that for a given set of incident conditions the collision-induced excess carrier density would be proportional to the optically-derived carrier recombination lifetime. Here, for different surface preparations we found different carrier recombination lifetimes. By measuring the collisionally excited carriers for several of these preparations, we were able to demonstrate this correlation. Fig. 4 shows a plot of the sample-averaged collisionally excited carrier density for Xe at 9.8 eV and normal incidence versus the carrier recombination lifetime measured at the same density for the same surface. Note that each data point corresponds to a different surface preparation.

3.3. Incident angle dependence of the yield

The excitation yield as a function of polar angle, θ_i, for Xe incident at 9.3 eV along three perpendicular azimuths, ϕ_i, is shown in fig. 5. The azimuths are measured with respect to the [001] direction as shown in the inset. Note that the excitation yield for the $\phi = 180°$ direction is nearly flat around $\theta_i = 0°$, but peaks off normal at approximately $\theta_i = 30°$. In contrast, the polar angle dependences for $\phi = 0°$, $\phi = 90°$, and $\phi = 270°$ are about the same, maximize at $\theta_i = 0°$, and are similar to those previously observed for InP(100) [3,5]. In each case the angle dependence is somewhat different than that expected if the excitation yield were proportional only to the average energy deposited, if it is assumed that the average energy deposited scales as $E_i \cos^2\theta_i$ (shown as the solid line in fig. 5).

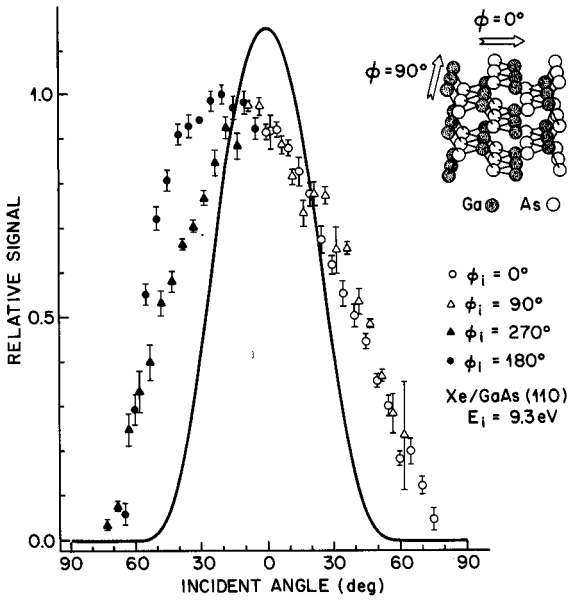

Fig. 5. Electron–hole pair excitation yield per incident atom versus polar and azimuthal angles (θ_i and ϕ_i, respectively) for Xe on GaAs(110) at an incident energy of $E_i = 9.3$ eV. The inset shows the various incident azimuthal angles for the GaAs(110) surface which are defined relative to the [001] direction as in ref. [8]. The solid line shows the expected distribution based on the slope of the line fit to the data of fig. 2, assuming that $\overline{\Delta E} = E_i \cos^2\theta_i$.

The striking difference in e^-h^+ excitation with azimuth, and the high absolute value of the e^-h^+ yield compared to the yield derived from the simple equilibration argument, raise the prospect that these results fall outside the regime of a transient equilibration between e^-h^+ excitations and the local impact region. We want to clearly note, however, that we do not know if the energy transfer from the Xe to the substrate maximizes at the same incident polar and azimuthal angles as the e^-h^+ yield, in which case the yield may still be exponentially proportional to the energy transfer as found for InP. We do not have the experimental data to give a direct comparison for the energy transfer near $\theta_i = 0°$ with the e^-h^+ yield. Based on the trends we observed for the energy transfer to GaAs(110) at more grazing angles of incidence [10,11], we do not think it likely that the energy transfer maximizes at the same angle of incidence as the e^-h^+ excitation yield. Further, the classical trajectory calculations, which successfully duplicate the scattered Xe distributions, indicate that the collision dynamics leading to the maximum energy transfer (backscattering) are similar at $\theta_i = 30°$ and $60°$ (for $\phi = 0°$). Thus we argue that the hyperthermal Xe–surface collision which results in the driving of a Ga atom into the bulk yields a more efficient excitation of e^-h^+ than a corresponding hit on an As atom. The higher excitation yield is a result of the collision dynamics (including the distortion of the electronic structure and the carrier escape probability) rather than simply depending on the increasing energy transfer.

Independent of the angular dependence, the estimated yield based on the local equilibrium argument is about a factor of 100 less than the experimentally derived yield, for which the fractional energy transfer of the incident Xe atom was assumed to be $> 80\%$. If the *total* incident beam energy (mean value) is assumed to be transferred, the increased yield based on the equilibration argument would still be over an order of magnitude less than the experimentally derived yield.

A generic picture of electronic excitation in atom–molecule collisions, in the absence of harpooning (electron jumps due to species with small ionization potentials colliding with species of large electron affinities), involves a collisionally induced distortion of the potential surfaces such that the ground and excited state surfaces approach. If the collision energy exceeds the electronic exitation gap, then upon separation the system may remain electronically excited. For convenience, one may consider three aspects of this problem separately: the electronic states in the unperturbed system that are mixed in the collisional distortion, the extent of their coupling during the collision, and the probability that the electron will remain in an excited state as the system relaxes. In an analogous fashion one can consider the electronic excitation at the GaAs(110) surface, involving distortions of the valence and conduction bands of GaAs. An additional aspect for the solid surface is the probability with which electrons (holes) leave the vicinity of the collision as carriers in the conduction (valence) band as the electronic states return and more smoothly connect to the undistorted bulk band structure.

The applicability of this simple picture is suggested by our understanding of the hyperthermal Xe and Kr collisions with the GaAs(110) surface. In our previous work [10] we have shown that hyperthermal collisions give rise to rainbow-like features (backscattered maxima) in the scattered projective angular distributions. Based on classical trajectory calculations [11] these features are understood to arise from nearly line-of-center impacts between the projectile and target surface atoms, followed by a rehitting of the projectile after the target rebounds from its nearest neighbors. These specific collisions lead to the maximum energy transfer and the greatest coordinate distortion of the lattice – two factors which should be of greatest significance in the collisional excitation of electron–hole pairs. We consider, therefore, that the dominant process leading to the excitation of e^-h^+ due to hyperthermal collisions is the severe distortion of the surface structure occurs when a single target atom is driven hard towards the bulk by a line-of-centers collision. Upon rebound from the bulk lattice, the rehitting of the incident projectile keeps the target atom from escaping and contains the energy transfer. In particular, with respect to the present experimental results, and appropriate model describing the excitations due to these collisions would indicate

what aspects of the GaAs(110) surface lead to a higher probability for electronic excitation and escape if the target atom is Ga rather an As.

4. Conclusions

We have measured the excitation probability of electron–hole pairs at the GaAs(110) surface due to impact of hyperthermal neutral atoms. The dependence on the angles of incidence and the high yield cast doubt on the appropriateness of an equilibrated local "hot-spot" description of the excitation process for this surface, although it was adequate to describe our previous results for InP surfaces. Thus these excitations on GaAs(110) may provide a basis for comparison and motivation to consider theoretically the dynamics of the collisional excitation of carriers at a semiconductor surface.

Acknowledgements

The authors would like to thank Ralph Logan for supplying the GaAs crystals, Murray Robins and Vince Lambrecht for evaporating contacts on them, Ed Chaban for assistance in setting up the experiments, and Greg Blonder, Frederico Capasso, Sue Coppersmith, Don Hamann, Michel Lannoo, Carmay Lim, Mark Stiles, and John Tully for helpful discussions. Also, the authors would like to thank Charlie Bahr for help in the preparation of the manuscript and figures.

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