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Picosecond Optical Measurements of Band-to-Band Auger Recombination of High-Density Plasmas in Germanium

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The recombination kinetics of transient high-density electron-hole plasmas in germanium has been measured by time-resolved free-carrier absorption with picosecond optical pulses. Density-dependent recombination rates as high as 4×10^9 sec^{-1} have been observed in plasmas with initial densities up to 3.4×10^{20} cm^{-3}. At this density the band-to-band Auger rate constant, $\gamma_3 = n^{-3}\,\partial n/\partial t$, is found to be 1.1×10^{-31} cm^6 sec^{-1} at room temperature in germanium.

At very high carrier densities in semiconductors, the third-order nonradiative band-to-band Auger effect can dominate all other recombination mechanisms such as radiative and surface recombination, and trapping at crystal defects. Theoretical estimates of the Auger rate constants and activation energies have been made for a number of materials, including InSb,[1] Ge,[2] Si,[2] and GaAs.[3,4] While experimental observations[5-8] have clearly demonstrated the existence of band-to-band Auger recombination, accurate determinations of the rate constants have proven to be more difficult to obtain.

Measurement techniques which have been used include the direct observation of decay tails of luminescence[8] and photoconductivity[5] following excitation by Q-switched lasers, and indirect measurements such as the intensity dependence

of photoconductive gain[7] and free-carrier absorption under quasi-steady-state conditions. The most complete results have been obtained with silicon, for which Blinov *et al.*[5] and Nilsson[8] found $\gamma_3 = 5\times10^{-30}$ and 4×10^{-30} cm^6 sec^{-1}, respectively, where the Auger rate constant, γ_3, is defined by the kinetic equation for the plasma recombination,

$$\partial n/\partial t = -\gamma_3 n^3. \quad (1)$$

The cubic dependence of the recombination rate arises from the three-body character of the Auger process, whereby an electron recombines with a hole, and the excess energy is transferred to another electron or hole in the form of kinetic energy. For germanium, Conradt and Aengenheister[7] have deduced an estimated rate constant of approximately 10^{-31} cm^6 sec^{-1} from an indirect measurement of the intensity dependence of the photoconductive response.

To measure Auger rate constants it is clearly desirable to work with very high-density plasmas in which the Auger recombination rate dominates all other competing processes such as radiative and surface recombination, diffusion, and trapping. Of these, diffusion is often the most severe, especially in those cases where the plasma is produced by optical excitation of a thin surface layer. Although Q-switched laser pulses of typically 10 to 30 nsec duration can produce high plasma densities, their time resolution is poor, obscuring the important initial features of the more rapid decays encountered at higher densities. They also tend to produce an undesirable amount of sample heating. A single picosecond pulse selected from a mode-locked pulse train can readily generate high plasma densities[9] with considerably less sample heating. The complete time-resolved Auger recombination can be observed, and the rate constant can be determined within a time interval which is less than typical diffusion times.

The particular application of picosecond optical pulses for the observation of Auger recombination in germanium is depicted schematically in Fig. 1. The technique utilized the excite and probe method, whereby the plasma was first generated in the crystal by band-to-band absorption of one pulse, and the kinetics of the plasma decay was subsequently measured by a second, longer-wavelength pulse using time-resolved free-carrier absorption. A mode-locked Nd:glass laser was used to produce pulses of approximately 8 psec duration and 10^8 W peak power at a

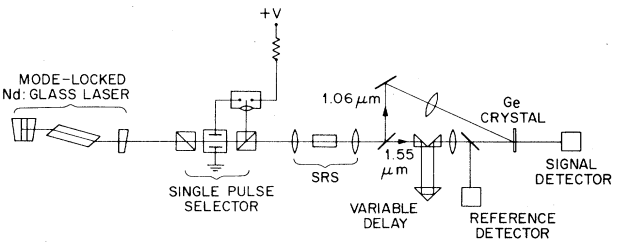

FIG. 1. Schematic diagram of experiment used for measurement of Auger recombination by time-resolved free-carrier absorption. The pump beam which generated the plasma was 1.06-μm pulse. A probe pulse at 1.55 μm was produced by stimulated Raman scattering.

wavelength of 1.06 μm. A single pulse was selected from the train by a laser-triggered spark gap and a Kerr cell. Focusing of the pump pulses at the crystal produced a measured energy density of 7.4×10^{-3} J/cm^2, a factor of approximately 2 below the damage threshold. To probe the plasma density by free-carrier absorption, a second pulse at 1.55 μm was produced by stimulated Raman scattering in benzene. Although this wavelength does not fall below the indirect gap in germanium, it is sufficiently long that band-to-band absorption (3.5×10^2 cm^{-1}) was negligible compared to the free-carrier absorption (typically 10^4 cm^{-1}). Tighter focusing of the probe pulses was used to achieve good overlap of pumping and probing pulses, and to ensure the probing of a moderately uniform plasma. The focal spot area of the probe was approximately one-fourth the pump spot area. A variable delay in the probe arm was used to adjust the timing of the free-carrier absorption measurement relative to the plasma generation. Germanium avalanche photodiodes, with thick silicon filters to block unwanted 1.06-μm radiation, were used to detect the incident and transmitted probe pulses. Analog-to-digital converters and a digital printer were used to record the data. Digital control of a stepping motor to vary the optical delay allowed completely automatic data collection, a feature which proved highly useful since averaging of as many as 25 laser shots for each delay were required to minimize statistical fluctuations in the data.

The germanium crystal was a (111) slice 0.3 mm thick having a room-temperature resistivity of 40 Ω cm, which was Syton polished on both faces. The linear absorption constant[10] at 1.06 μm is 1.4×10^4 cm^{-1}. The linearity of the absorption was tested in a separate crystal 3 μm thick, and no appreciable deviation from linearity was

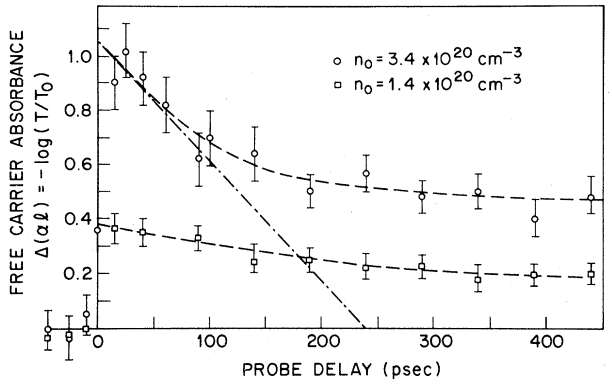

FIG. 2. Time evolution of the plasma obtained from free-carrier absorption versus optical delay time between pumping and probing pulses. Error bars represent statistical variance of data.

observed up to the intensities encountered in this experiment.

The results of the Auger measurement are shown in Fig. 2, where the free-carrier absorbance of the plasma at 1.55 μm is plotted versus the time delay following plasma generation by the 1.06-μm pulse. The absorbance is obtained from the logarithm of the transmission ratio of the probe beam and is proportional to the integrated plasma density though the crystal (the probe transmission before plasma generation, T_0, was approximately 4%). Recombination curves are shown for two different initial plasma densities. The initial plasma density was determined from a direct measurement of the incident photon flux with a calibrated detector. This measurement was made through a small pinhole located at the focus of the pump beam, eliminating the need to measure the shape and size of the focal spot. The incident pump energy density was found to be 7.4×10^{-3} J/cm^2 or 4×10^{16} photons/cm^2, which produced an initial plasma density of 3.4×10^{20} cm^{-3} at the crystal surface, under the assumption of an exponential absorption depth of 7.1×10^{-5} cm. The initial plasma density can also be deduced from the magnitude of the free-carrier absorption. Using $\sigma_p = 3.3\times10^{-17}$ cm^2 and $\sigma_n = 4\times10^{-18}$ cm^2 for the hole and electron free-carrier cross sections,[11] respectively, and the observed absorbance of 1.4×10^4 cm^{-1}, the initial density was 3.8×10^{20} cm^{-3}, in good agreement with the direct measurements of photon flux.

The results for the reduced initial plasma density were obtained by attenuating the pump by a factor of 0.4 with a neutral density filter. The reduction in free-carrier absorbance by the same amount indicates that the cross sections are independent of density within the range of observation.

From previous measurements by picosecond ellipsometry[9] of the diffusion of high-density plasmas in germanium, we expect the initial plasma layer to double its effective thickness in approximately 75 psec. In the absence of recombination, the free-carrier absorption would be unaffected since diffusion conserves the total number of particles (the diffusion transverse to the probe beam is negligible). However, since the Auger decay rate depends on the cube of plasma density, the overall behavior of the decay of the free-carrier absorption will be affected by diffusion because of the redistribution of carriers. From an analytical point of view, the behavior is rather complicated, requiring the numerical solution of a nonlinear partial differential equation. However, in the early phases of decay (i.e., first 75 psec), the kinetics is dominated by the Auger recombination, and is independent of diffusion to first order. More specifically the initial decay rate is

$$\frac{(\partial/\partial t)[\int_0^L(\sigma_n+\sigma_p)n(z,t)\,dz]_{t=0^+}}{\int_0^L(\sigma_n+\sigma_p)n(z,t)\,dz} = \frac{1}{3}\gamma_3 n_0^2 \qquad (2)$$

for the case of an exponential spatial distribution $n(z,0) = n_0 e^{-\alpha z}$, under the assumption that the crystal thickness L is much greater than the pump absorption depth, α^{-1}. The value of the initial decay rate is shown as a dotted line in Fig. 2, and is equal to 4×10^9 sec^{-1} for $n_0 = 3.4\times10^{20}$, producing a value of the rate constant $\gamma_3 = 1.1\times10^{-31}$ cm^6 sec^{-1}. This result is in good agreement with the result of Conradt and Aengenheister.[7] At this density the radiative decay rate is only 1.5×10^7 sec^{-1}, and contributes less than 1% to the total recombination rate. Surface recombination and trapping are expected to be unimportant on this time scale.

Huldt[2] has made a theoretical estimate of the Auger rate constant for Ge at room temperature for nondegenerate plasmas. He finds $\gamma_3 = 7\times10^{-33}$ cm^6 sec^{-1}, which is a factor of 15 smaller than our experimental result. We would not expect good agreement, however, since at the densities in our experiment, the plasma is clearly degenerate, and also the electron and hole Fermi energies exceed the activation energy which he estimates to be at least 0.16 eV. In the range of very high densities, the degenerate property of

the plasma may also contribute to a reduction in the Auger recombination rate as a result of the action of the Pauli exclusion principle in limiting the number of available final states. It is possible that this latter effect is evident in the fact that at the reduced (but still degenerate) density of 1.4×10^{20} cm^{-3}, we find that the rate constant is larger by a factor of approximately 1.8 than at 3.4×10^{20} cm^{-3} (Fig. 2).

In summary, we have demonstrated the utility of picosecond optical pulses for the measurement of fast nonradiative band-to-band Auger recombination of high-density semiconductor plasmas. The high intensity and short duration of picosecond pulses allows the study of recombination kinetics in a density and time regime where the Auger effect dominates all other recombination processes. Furthermore, it is possible to determine the rate constant in a time interval which is short compared to diffusion times.

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Angle and Energy Dependence of Photoemission from NaCl and KCl Single Crystals*

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The dependence of photoemission on the azimuthal emission angle at fixed polar angle of 45° was measured for the (100) face of NaCl and KCl single crystal cleaved *in situ*. The initial- and final-state energy was scanned using synchrotron radiation. Strong variations of the angular pattern are observed for different final-state energies. Below the electron-electron scattering threshold the pattern depends only on the final-state energy. Electron-phonon scattering can account for this fact.

Experimental data suitable for testing band-structure calculations for alkali halides are remarkably scarce in view of the enormous number of experiments which have been performed with these fundamental compounds. The reason for this is the dominating influence of excitons on the optical spectra and the excellent insulating properties which, so far, have restricted photoelectron spectroscopy to samples of thin evaporated films.

In this Letter we report on the first ultraviolet photoemission experiment with bulk alkali halide single crystals. This allows us to measure the angular distribution of the photoelectrons, as has been done before with metals and semiconductors.[1-7] Although our investigation was incomplete since the polar angle could not be varied, the data yield considerably more detailed information than former experiments which integrated over all angles. They show that angle-resolved photoelectron spectroscopy on alkali halides is possible and may prove to be a promising method of investigating the electronic structure in detail, if the problem of matching the wave functions at the surface can be attacked successfully.[8]

We have used the synchrotron radiation of DESY as a light source. This enabled us to vary the photon energy continuously and to scan either the initial- or the final-state energy of the photoelectrons. The experimental equipment has been described prevously.[9,10] It consisted essentially of a normal-incidence monochromator with 2-Å resolution and an ultrahigh-vacuum sample chamber with a base pressure in the 10^{-11}-Torr range.