Intensity of exciton luminescence in silicon in a weak magnetic field

W. M. Chen, O. O. Awadelkarim, H. Weman,* and B. Monemar†
Department of Physics and Measurement Technology, Linköping University, S-581 83 Linköping, Sweden
(Received 9 April 1990)

The effect of a weak magnetic field on the intensity of photoluminescence (PL) due to exciton recombination in silicon is investigated. A considerable decrease (as much as 10–40 % for a rather low optical excitation level, <1 W/cm^2) in the PL intensity with above-band-gap excitation is observed with increasing magnetic field (<0.25 T). The magnitude of this change depends on optical pumping level, excitation photon energy, orientation of the crystal with respect to the magnetic field, as well as the near-surface quality of the crystal. The mechanism responsible for this phenomenon is discussed and is attributed mainly to a strong enhancement of surface recombination due to magnetic-field-induced confinement of photoexcited free carriers near the surface.

I. INTRODUCTION

The effect of an external magnetic field on the luminescence intensity in solids has received great attention in the past. Magnetic-field-induced resonant-like changes in the luminescence intensity have been extensively investigated and are quite well understood. Such changes were mostly attributed to cross relaxation (CR), an important mechanism for intercenter energy transfer, and also to level-anticrossing (LAC) effects of an intracenter nature.[1–3] The nonresonant variations of the luminescence intensity in a magnetic field, in contrast, are not yet as well understood, although these variations are commonly observed as background signals in a magneto-optical spectrum.

Investigations of such a magnetic-field-induced change in photoluminescence (PL) intensity (denoted as MFIC-PL below for brevity) due to electron-hole droplets (EHD) in germanium have been carried out,[4,5] both with bulk excitation and with surface excitation.[6] Similar studies have also been done for free excitons (FE's) in germanium.[7,8] In this paper we present an investigation of PL intensity due to both FE and bound-exciton (BE) recombinations in silicon in the presence of a weak static magnetic field. The mechanism responsible for the magnetic-field effect will be discussed in detail.

II. EXPERIMENT

A variety of silicon crystals were used in this work, grown by the float zone (FZ) or Czochralski (Cz) method, undoped or lightly doped. Some of them were also treated by a low-dose electron irradiation ($\sim 1.0\times10^{16}$ e/cm^2) and/or sequential thermal annealing ($\sim$400 °C). The cut samples were then stuck to a jig using adhesive black wax and thinned to a desired thickness of $\sim$1.0 mm by being lapped on a glass flat. In the thinning process, silicon carbide powder in the grain size range 240–800 mesh was used. The samples were chemically cleaned using trichloroethylene and then polished in a Kent MK2A machine using a diamond compound (Hyprez 5-star) in four different grades with the appropriate polishing disks. After the polishing procedure possible surface damage and adsorbed foreign atoms were removed using a solution of hydrofluoric acid. The sample surface was then cleaned by rinsing the sample in ethanol, acetone, and distilled water.

PL signals emitted from the samples at 2 K were collected by a liquid-nitrogen-cooled North Coast EO-817 Ge detector via a Jobin-Yvon 0.25 m grating monochromator. The magnetic-field dependence of the PL intensity was obtained with the aid of a magnet and a time base (in continuous scanning mode) attached to a modified Bruker 200D-SRC ESR spectrometer, accessible up to 1.5 T. Above-band-gap optical excitation was provided by the 5145 Å line of an Ar^+ laser or a 7500 Å line from a dye laser (with Styryl 9 dye), working at a low pumping level with an unfocussed laser spot (<1 W/cm^2) to avoid possible condensation of excitons to EHD's and local heating. The high-resolution PL spectrum was obtained with the aid of a Spex-1404 0.85-m double-grating monochromator.

III. RESULTS AND DISCUSSION

The effect of a magnetic field on the intensity of FE and BE luminescence is observed to be similar for all samples under study, though quantitatively different from sample to sample. The physical mechanism responsible for the effect is the same, however. Below we shall only show as examples the experimental results obtained from two FZ silicon samples: sample 1 is a pure one (with resistivity $\sim$10 000 Ω cm), and sample 2 is lightly boron doped (100 Ω cm) after e irradiation.

In Fig. 1 we show low-temperature PL spectra from such samples. In sample 1 the TO-phonon-assisted FE transition is dominant in the PL spectrum, together with the B-related shallow BE emission (denoted as B-BE below). However, the PL spectrum of sample 2 is dominated by the isoelectronic BE recombination, the G line.[9]

In Fig. 2 we show the magnetic-field dependence of the FE PL intensity from sample 1 and BE's (B-BE and G line) recombinations from sample 2 at two excitation lev-

els ($I_1 \approx 5I_2$). All of the emissions display a marked variation in the intensity with increasing magnetic-field strength. In addition, a stronger variation is obtained in the case of the lower excitation level than in the case of the higher excitation level. The different line shape for the FE and the BE's is closely related to the efficiency of the exciton formation with respect to the decay, as will be discussed in more detail below.

In Fig. 3, we show the MFIC-PL spectra of the FE and the B-BE obtained from both the polished and unpolished surfaces of the sample, with the same excitation level and crystallographic orientation in both cases. A stronger variation in the PL intensity with the magnetic field is observed for the light emitted from the unpolished surface of the sample in comparison with that emitted from the polished surface. This implies a surface-dependent nature of the MFIC-PL spectra.

In Fig. 4 we demonstrate an angular dependence of the MFIC-PL spectra for the B-BE. A stronger variation is observed when the magnetic field is at an angle $\sim 75°$ to the normal direction of the sample surface, compared to the case of $\sim 35°$.

To account for the excitation, surface, and angular dependences of the experimental MFIC-PL spectra, we shall now examine the possible effects of the external static magnetic field on the exciton (FE and BE) recombination.

First of all, a magnetic field leads to a transverse compression of the ground-state wave function of the exciton,[7,8] and consequently intensifies the annihilation of the exciton; the amplitude for the annihilation of the exciton is proportional to its density at the origin of the relative motion of the electron and hole in the exciton (i.e.,

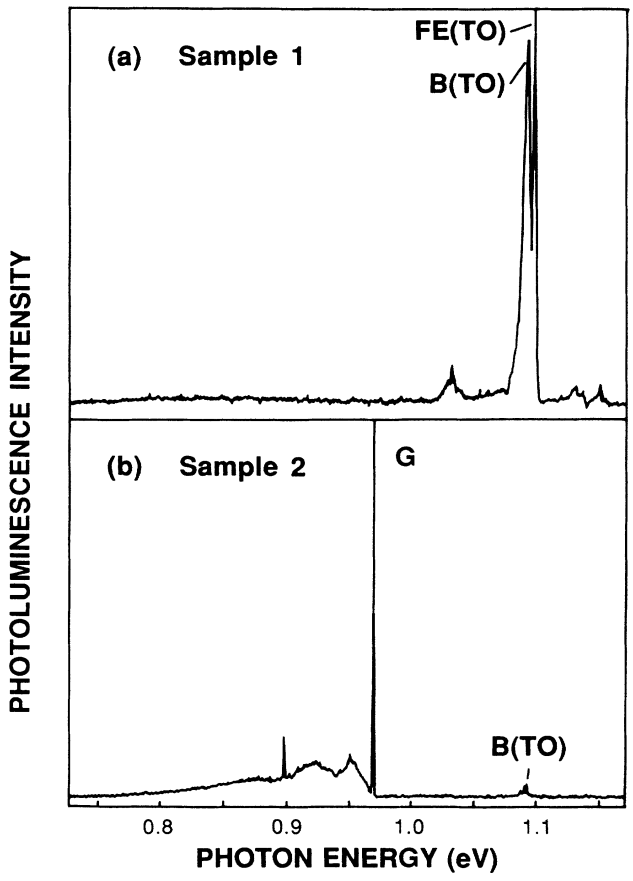

FIG. 1. Photoluminescence spectra on a linear scale from (a) sample 1 and (b) sample 2, at 2 K. FE(TO) denotes the transverse optical-phonon-assisted free-exciton transition, and B(TO) that for the neutral boron-acceptor bound exciton (BE). The *G* line arises from the isoelectronic BE recombination (Ref. 9).

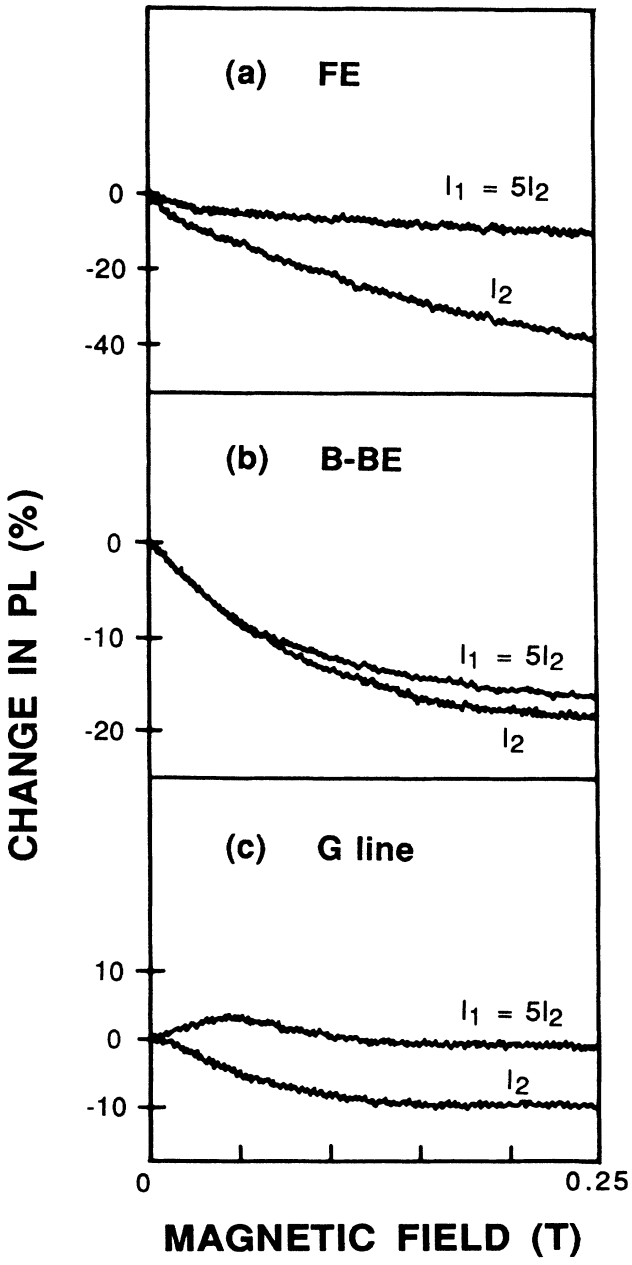

FIG. 2. magnetic-field-induced change in photoluminescence (MFIC-PL) spectra from (a) the free exciton, (b) the shallow B-related BE, and (c) the *G* line BE in silicon, at two excitation levels ($I_1 \approx 5I_2$). I_1 corresponds to an excitation level of ~ 1 W/cm^2.

to the strength of overlap between the electron and hole wave functions). The above is certainly suggestive of an increase rather than the *decrease* in the FE recombination observed in this work. Moreover, the magnetic Stark effect,[10] which affects mainly the forbidden transitions, is expected to give rise to an *increase* in PL intensity due to the effect of the magnetic field on the motion of the exciton.[8]

A perturbation by a magnetic field can also cause a redistribution of excitons among spin sublevels and a change in radiative recombination rates for the FE and the BE states due to magnetic-field-induced mixing of wave functions, i.e., an effect of symmetry breaking. This may result in a change in the PL intensity. However, this bulk effect cannot explain the surface dependence, nor the angular dependence in the case of the FE, since the FE states are isotropic when the tetrahedral crystal field is negligible, which is the case here. The influence of possible magnetic-field-dependent recombination channels or trapping of free carriers and excitons can also be excluded for the same reason.

In the studies of the EHD luminescence intensity in germanium, a change in the quantum efficiency of the luminescence emitted from electron-hole drops was observed in a magnetic field, and was explained as being due to the recapture of some of the Auger particles emitted from the drops.[5] The angular dependence was shown there to result from the anisotropy of the phonon wind,[11] which is the driving force behind the motion of the EHD's in the crystal.[12–14] However, this explanation was questioned by the kinetic study of the relaxation of the EHD luminescence, as pointed out in Ref. 4. This effect on the EHD's is believed to be insignificant for the case of excitons here.

Since the bulk effects of the magnetic field on the exciton luminescence could not explain our experimental results, a possible mechanism may then be related to surface recombination. Such a mechanism is plausible because the surface recombination plays an important role under our experimental condition with above-band-gap excitation, especially by the 5145 Å line, in which case the depth of penetration of the light into the silicon crystal [$\alpha^{-1} \approx 1$ μm (Ref. 15)] is much less than the diffusion lengths of the photogenerated carriers [$L_e > 100$ μm (Ref. 16)]. In what follows, we shall show that this explanation is relevant and consistent with all of our experimental observations.

Let us now consider the behavior of a system of nonequilibrium carriers and excitons in an applied magnetic

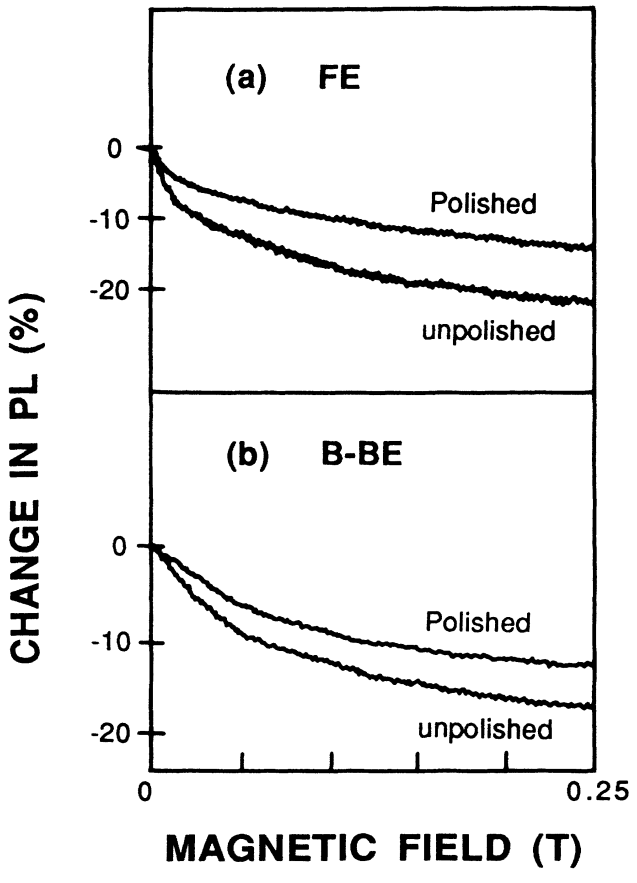

FIG. 3. MFIC-PL spectra from (a) the FE and (b) the B-BE, obtained from the polished and unpolished surfaces of the sample with the same excitation level and crystallographic orientation ⟨110⟩.

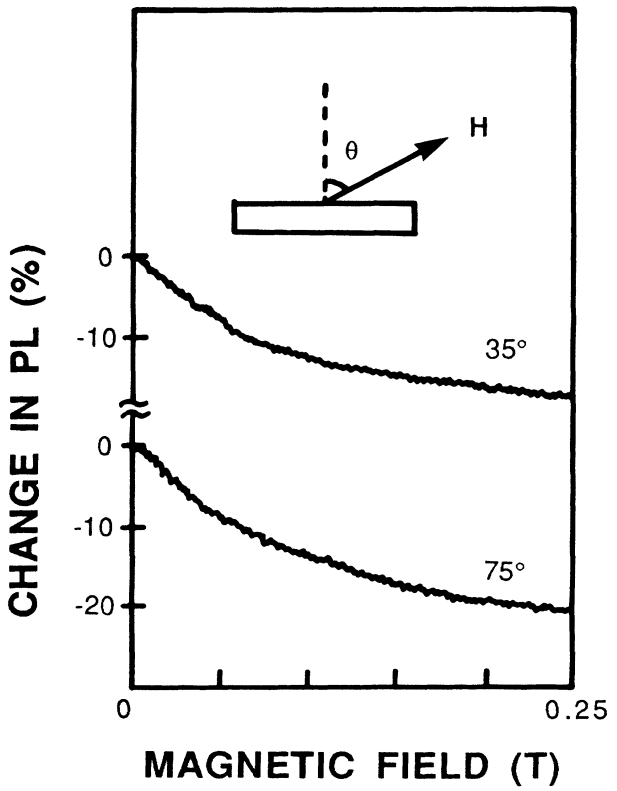

FIG. 4. MFIC-PL spectra from the B-BE, when the magnetic field is at an angle $\theta \sim 75°$ and $\theta \sim 35°$ to the normal direction of the sample surface. The inset shows the definition of the angle θ.

field. The continuity equations for the system have the form

$$D_e\frac{d^2n_e}{dx^2}+G-\gamma n_e^2+\beta n_{ex}-\frac{n_e}{\tau_e}=0\ ,$$
$$D_{ex}\frac{d^2n_{ex}}{dx^2}+\gamma n_e^2-\beta n_{ex}-\frac{n_{ex}}{\tau_{ex}}=0\ . \tag{1}$$

Here, $G=\alpha I_0 e^{-\alpha x}$ is the carrier generation rate and τ_e and τ_{ex} are the respective lifetimes for the free carriers and the excitons. γ is the coefficient of binding electrons and holes into excitons. β is the thermal dissociation coefficient, which is believed to be unimportant in our low-temperature measurement (2 K). D_e and D_{ex} are the coefficients representing diffusion of the carriers and the excitons, respectively.

By solving Eq. (1) with the proper boundary conditions in the surface excitation case,[4] we can obtain the density distribution functions for both the free carriers and the excitons and their concentrations as a function of magnetic field. The magnetic-field dependence of the exciton PL intensity can be consequently obtained, knowing that the PL intensity is proportional to the corresponding exciton density, i.e.,

$$I\sim k^r n_{ex}\ , \tag{2}$$

where k^r is the radiative decay rate of the exciton. Such dependences are shown in Figs. 5 and 6 for two extreme cases: (1) $\gamma n_e^2 \ll n_e/\tau_e$—the recombination of the free carriers is much more efficient than the binding of the free carriers into the excitons; (2) $\gamma n_e^2 \gg n_e/\tau_e$—the binding into the excitons is dominating over the recombination of the photogenerated free carriers. In Figs. 5(a) and 6(a) we show the calculated MFIC-PL spectra with varying excitation level. It is concluded that the MFIC-PL spectrum is rather strongly dependent on the excitation level in the second case, while this is not true in the first case. In both cases the MFIC-PL spectrum varies significantly with the quality of the sample surface (i.e., the surface recombination rate s) and with the excitation photon energy (i.e., the penetration of the excitation light with respect to the carrier diffusion length). The two cases differ from each other, however, in the MFIC-PL

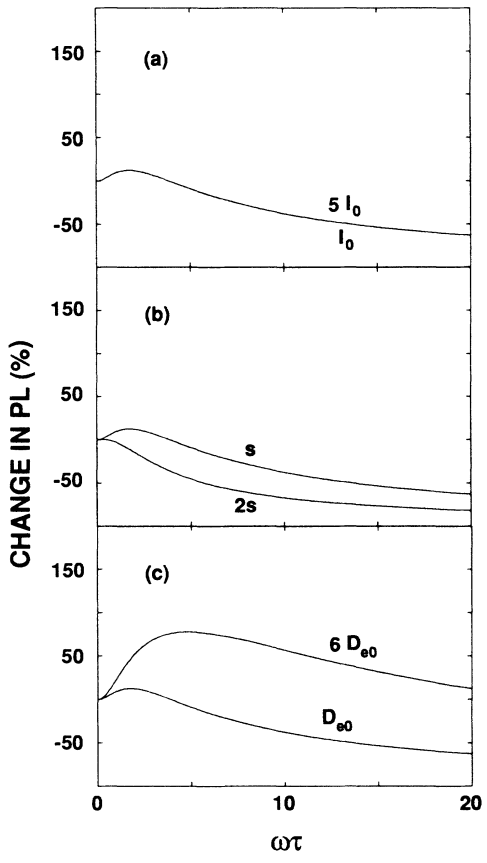

FIG. 5. MFIC-PL spectra, calculated according to Eqs. (1) and (2) in the case $\gamma n_e^2 \ll n_e/\tau_e$, as a function of (a) the excitation rate I_0, (b) of the surface recombination rate s, and (c) of the zero-field carrier diffusion length D_{e0}. The specific parameter values used are $I_0=2\times10^{17}$ cm^{-2}sec^{-1}, $s=5\times10^3$ cm/sec, $D_{e0}=1\times10^2$ cm^2/sec, $\gamma=1\times10^{-4}$ cm^3/sec, and $\tau_e=1\times10^{-6}$ sec.

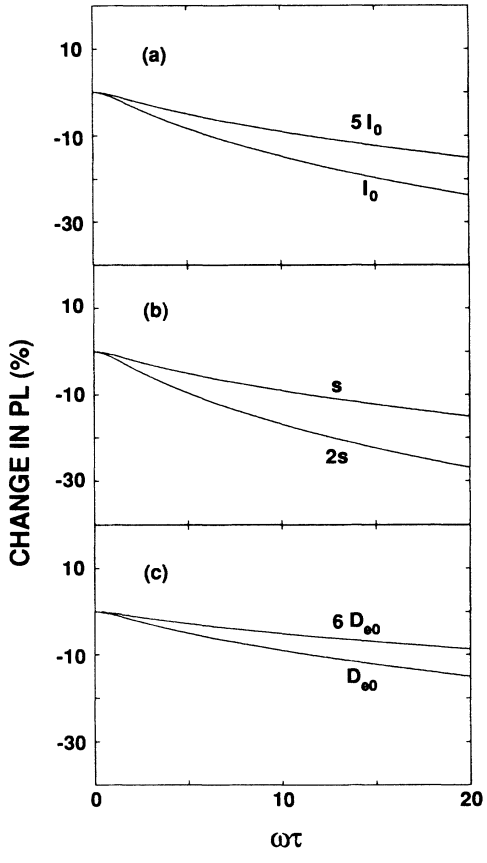

FIG. 6. MFIC-PL spectra, calculated according to Eqs. (1) and (2) in the case $\gamma n_e^2 \gg n_e/\tau_e$, as a function of (a) the excitation rate I_0, (b) of the surface recombination rate s, and (c) of the zero-field carrier diffusion length D_{e0}. The specific parameter values used are $I_0=2\times10^{17}$ cm^{-2}sec^{-1}, $s=5\times10^3$ cm/sec, $D_{e0}=1\times10^2$ cm^2/sec, $\gamma=1\times10^{-4}$ cm^3/sec, and $\tau_e=1\times10^{-6}$ sec.

spectral line shape. In the first case, the PL intensity increases at lower fields and then decreases. Whereas in the second case only a monotonous decrease in the PL intensity is expected with increasing magnetic field.

Our observations for the FE and the shallow B-related BE emissions in silicon crystals seem to indicate that the second case mentioned above is valid, which is consistent with very efficient binding of the free carriers into the FE and the shallow B-BE. The G line BE, however, may be viewed as an intermediate case. A quantitative analysis of the MFIC-PL spectra was not attempted in this work due to problems of ununiqueness of the fitting parameters, which requires an independent determination of some of the parameters by other complementary techniques.

The observed MFIC-PL is a direct result of strongly enhanced surface recombination due to magnetic-field-induced confinement of the photogenerated free carriers near the sample surface. This magnetization is mainly reflected by the magnetic-field dependence of the diffusion coefficient of the carriers by $D_e(H)=D_0/(1+\omega_H^2\tau^2)$, where $\omega_H=eH_\perp/m^*c$ [$H_\perp=H\sin\theta$ is the magnetic-field component perpendicular to the direction of diffusion of the carriers (Fig. 4)]. This explains the angular dependence of the MFIC-PL spectra studied in the present work: a stronger variation is observed when the magnetic field is at an angle 75° to the normal direction of the measured sample surface in comparison with the case of 35°, due to a higher confinement of the carriers near the surface in the former case. Actually, it is possible to obtain an equivalence between these two cases if the effective strength of the magnetic field inducing the confinement (i.e., the transverse component of the magnetic field, $H_\perp$) is taken into consideration, as shown in Fig. 7.

Since the MFIC-PL spectra are directly related to the profile of the carrier indiffusion, which is determined by the photogeneration and the surface-recombination velocity of the nonequilibrium carriers, it is not difficult to understand why they should relate to the quality of the sample surface, as observed in the experiments. The dependence of the MFIC-PL spectra for the excitons on the excitation rates can also be explained in the same manner, in agreement with theoretical expectations (Fig. 6). Further support is given by employing optical excitation with different photon energy (7500 Å here), where a slightly weaker magnetic-field dependence is observed (Fig. 8), in agreement with a slightly deeper penetration of the light [$\alpha^{-1}\approx 6\,\mu$m (Ref. 15)].

The model discussed above, in terms of the high surface recombination, is valid as long as the surface excitation is considered, namely, the diffusion length of the photogenerated carriers is much larger than the penetration depth of light into the crystal. The opposite situa-

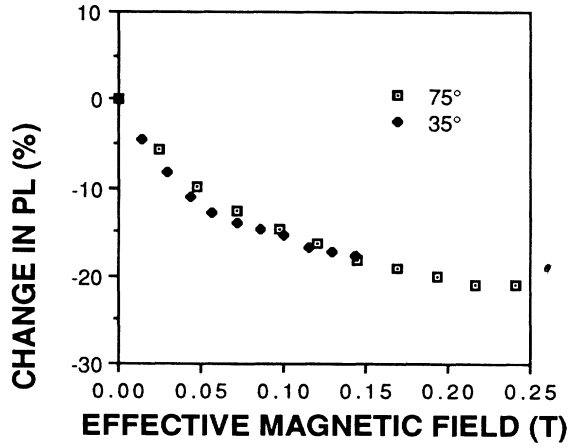

FIG. 7. MFIC-PL spectra from Fig. 4, where the horizontal axis is the effective magnetic field $H_\perp$, i.e., the magnetic-field component perpendicular to the direction of diffusion of the carriers.

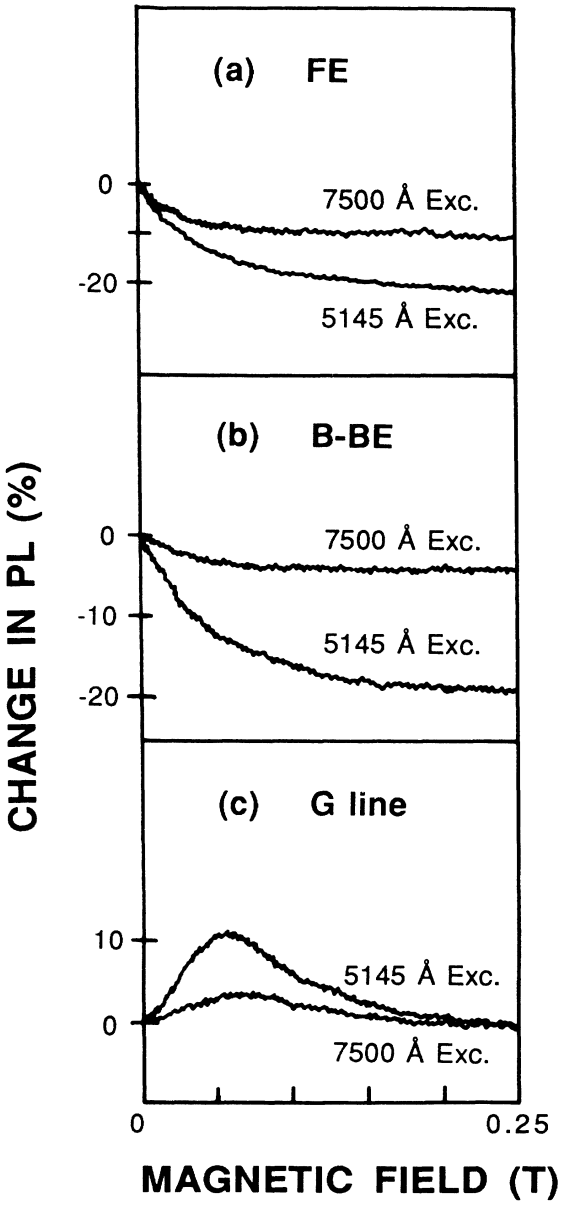

FIG. 8. MFIC-PL spectra from (a) the FE, (b) the B-BE, and (c) the G line BE in silicon, at two different excitation wavelengths, 5145 and 7500 Å, respectively.

tion, i.e., the bulk excitation (or bulk carrier generation), will give a very different picture of the condensation of the free carriers into excitons in a nonequilibrium system. The surface recombination no longer plays an important role in the intensity of the exciton luminescence in this case, as can be expected. A monotonous increase in exciton luminescence in germanium was observed with increasing magnetic field, due to the shrinkage of the orbit of relative motion of electron and hole in the exciton.[7] This case has not been experimentally studied in this work.

IV. CONCLUSIONS

We have investigated the effect of a weak magnetic field on the PL intensity of both the free- and the bound-exciton recombination in silicon, under the surface excitation condition. A considerable variation of the intensity of the exciton luminescence was consistently observed with increasing external magnetic field. The dependences on the excitation rate and the excitation photon energy, the transverse magnetic-field strength, as well as the quality of the crystals near the surface were also observed. Various possible effects of the weak magnetic field on the properties of the exciton luminescence processes were discussed. It was concluded that the main mechanism responsible for the experimental observations in the present work is due to the strong enhancement of the surface recombination by the magnetic-field-induced confinement of the nonequilibrium carriers near the sample surface. A detailed comparison with the theoretical model may in turn yield valuable information about the mechanism of exciton formation as well as transport properties of the material. The effect observed in this work indicates that caution must be exercised in the line-shape analysis of magneto-optical spectra to obtain electronic and transport properties of the materials studied. This is particularly true for studies of the influence of magnetic field on the exciton wave functions by detecting the variation in the exciton PL intensity (such as the LAC spectroscopy) and on the ODCR (optical detection of cyclotron resonance) spectrum in solids, where possible contribution from the surface effect has so far often been ignored.

ACKNOWLEDGMENTS

We are grateful to A. Henry for kind help and discussion in connection with the high-resolution PL measurements.

---

*Present address: Center for Quantizied Electronic Structure, University of California, Santa Barbara, CA 93106.

†Present address: Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, D-7000 Stuttgart 80, Federal Republic of Germany.

[1]M. Goldman, *Spin Temperature and Nuclear Magnetic Resonance in Solids* (Oxford University Press, London, 1970).

[2]For a review, see, e.g., D. H. Levy, in *Advances in Magnetic Resonance,* edited by J. S. Wangh (Academic, New York, 1973), Vol. 6, p. 1.

[3]W. M. Chen, M. Godlewski, B. Monemar, and P. Bergman, Phys. Rev. B **41**, 5746 (1990).

[4]B. M. Ashkinadze and V. V. Bel'kov, Fiz. Tverd. Tela (Leningrad) **26**, 974 (1984) [Sov. Phys.—Solid State **26**, 595 (1984)].

[5]D. Bimberg and M. S. Skolnick, Solid State Commun. **32**, 1311 (1979).

[6]By surface excitation, we mean that the depth of penetration of the excitation light into the crystal is much shorter than the diffusion length of the photogenerated free carriers. The opposite case will correspond to bulk excitation.

[7]V. D. Bisti, V. M. Edel'stein, I. V. Kukushkin, and V. D. Kulakovskii, Solid State Commun. **44**, 197 (1982).

[8]V. D. Kulakovskii and V. M. Edel'stein, Zh. Eksp. Teor. Fiz. **86**, 338 (1984) [Sov. Phys.—JETP **59**, 195 (1984)].

[9]A. V. Yukhnevich, Fiz. Tverd. Tela (Leningrad) **7**, 322 (1965) [Sov. Phys.—Solid State **7**, 259 (1965)].

[10]J. J. Hopfield and D. G. Thomas, Phys. Rev. **122**, 35 (1961).

[11]L. V. Keldysh, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 100 (1976) [JETP Lett. **23**, 86 (1976)].

[12]J. C. Hensel and R. C. Dynes, Phys. Rev. Lett. **39**, 969 (1977).

[13]J. Doehler and J. M. Worlock, Phys. Rev. Lett. **41**, 980 (1978).

[14]M. Greenstein and J. P. Wolfe, Phys. Rev. Lett. **41**, 715 (1978).

[15]G. E. Jellison and F. A. Modine, Appl. Phys. Lett. **15**, 180 (1982).

[16]M. Saritas and H. D. McKell, J. Appl. Phys. **63**, 4561 (1988).