Optical Detection of Cyclotron Resonance in Semiconductors

R. Romestain
Laboratoire de Spectrométrie Physique, F-38041 Grenoble, France

and

C. Weisbuch[a]
Laboratoire de Physique de la Matière Condensée, Ecole Polytechnique, F-91120 Palaiseau, France
(Received 4 August 1980)

The first optical detection of microwave cyclotron resonance via band-gap luminescence is reported. The method is based on the measurement of resonant heating of the carriers by monitoring shapes and intensities of low-temperature photoluminescence lines. As an example, electron and hole resonances in GaAs and CdTe are studied. Mobilities of up to 1.9×10^6 cm^2 V^{-1} s^{-1} have been observed. The method should be applicable to a wide range of compounds and alloys.

PACS numbers: 76.40.+b, 71.25.Jd, 78.20.Ls

The determination of the Luttinger parameters of the valence and conduction bands of semiconductors usually relies on two types of experiments, magneto-optics of interband transitions[1] and cyclotron resonance[2] (CR). In this Letter we demonstrate the feasibility of an optical detection of CR, which like the well-known optical detection of magnetic resonance,[3] appears to be very sensitive. Optical excitation of carriers by light exceeding the gap energy allows to detect CR on both types of carriers by the following mechanism: The absorption of microwave power occurring at the CR of either type of carrier increases the energy of both the electron and hole gases. The resulting carrier heating can be observed directly by measuring the effective temperature via the shape of optical emission lines or indirectly by monitoring luminescence line intensities. The measured values of the hole masses in GaAs and electron masses in CdTe and GaAs are in good agreement with standard CR measurements. The mobilities deduced from the cyclotron linewidth are the highest reported up to now for these compounds.[4] This is due to the neutralization of ionized impurities by photoexcited carriers. Finally, we obtain a first value for the (110) hole masses in CdTe: $m_{lh}=(0.12\pm 0.02)m_0$ and $m_{hh}=(0.81\pm 0.05)m_0$.

One might wonder why it is useful to develop a new method of detection of cyclotron resonance in semiconductors more than twenty-five years after its first observation.[2] Microwave CR has been very successful as an experimental tool, in particular for the detailed analysis of the band extrema in Ge[5] and InSb.[6] However, only few semiconductors have been measured by this technique, either for electrons or holes, seldom for both. This is largely due to the constraints of the microwave detection, which requires large monocrystals having low conductivity to maintain good cavity qualities. The optical excitation and detection scheme has several advantages over the standard CR scheme: The condition $\omega_c\tau \geqslant 1$ is satisfied at lower fields because of the light-induced impurity neutralization. In this way, the field dependence of the cyclotron-resonance line shape, which is still a matter of controversy[1,5] can be studied in detail over an extended range of magnetic field. In particular, the low-field limit where standard methods cease to fulfill the $\omega_c\tau$ requirement becomes accessible. Also, the optical scheme allows to investigate spotlight-size samples, single crystallites of polycrystalline material or inhomogeneous samples like epilayer alloys. Impurity distribution can be tested via mobility variations down to a scale unattainable by standard transport measurements. Finally, the new scheme should provide new insights in microwave-carrier-heating studies thanks to the direct determination of the carrier distribution functions in the same way as optical studies of hot electrons[7,8] have revived dc-carrier-heating studies.

To perform the experiment, high-purity GaAs or CdTe samples are mounted in a cylindral microwave cavity (TE_{013} mode) which is immersed in superfluid helium ($T=1.5$ K). The maximum microwave power at 70 GHz is $\simeq$200 mW (0 dB). A superconducting magnet creates the dc magnetic field. Free carriers are photoexcited by the light from an Ar-ion laser focused onto the sample through a small hole in the cavity. The photoluminescence light emerging from the sample is analyzed with a 0.75 m grating spectrometer

© 1980 The American Physical Society

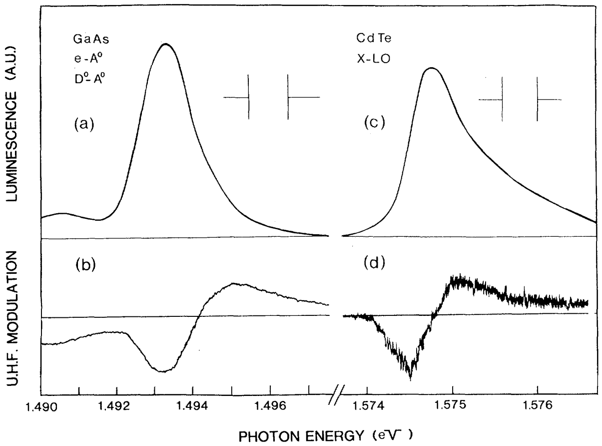

FIG. 1. Photoluminescence [(a), (c)] and its microwave-induced changes at cyclotron resonance of electrons [(b), (d)]: (a) Electron-acceptor ($h\nu \sim 1.493$ eV) and donor-acceptor ($h\nu \sim 1.490$ eV) recombination bands in GaAs. (b) Lock-in detected intensity change induced by a square-wave (on-off) modulation of the microwave power, the magnetic field being set at the electron CR (microwave power: -15 dB). Note the sign inversion in the intensity of the e-A^0 band, characteristic of an effective temperature change according to the line-shape function. (c) LO-phonon–assisted recombination of free excitons in CdTe. (d) Same as (b) on the LO-phonon line (-15 dB).

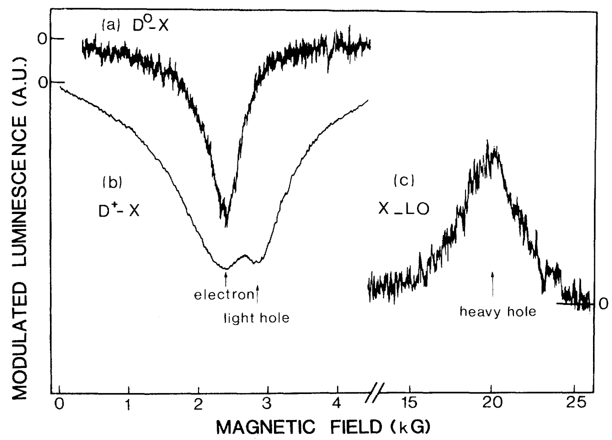

FIG. 2. Cyclotron resonance detection in CdTe: (a) D^0-X recombination line (1.5938 eV) only shows the electron CR (-20 dB). (b) D^+-X recombination line (1.5925 eV) shows electron and hole resonances (only the low field region is shown, displaying the electron and light-hole resonances). Because of the proximity of the two CR lines, the light-hole mass determination is unprecise (0 dB). (c) X-LO line at 1.576 eV showing the heavy-hole CR (note the signal sign corresponding to an intensity increase) (0 dB).

equipped with a GaAs-cathode cooled photomultiplier. Measurements were made at low light intensities ($I < 300$ mW/cm^2) to minimize line broadening due to carrier-carrier collisions.

When the CR condition $\omega_c = eB/m^*$ (where m^* stands for either the electron, light-hole, or heavy-hole effective mass) is fulfilled, resonant absorption of microwave energy occurs. Under our optical excitation conditions ($I \sim 100$ mW/cm^2), this excess energy is distributed over the carrier gases through electron-electron, electron-hole, and hole-hole collisions.[7] The carrier effective temperatures are thus resonantly increased, influencing photoluminescence lines. The change in carrier temperature is directly evidenced on the electron-to-acceptor line e-A^0,[7] whose line shape is given by

$$f(h\nu) = [h\nu - (E_G - E_A)]^{1/2} \exp[-(h\nu - E_G + E_A)/kT_e].$$

Here $h\nu$ is the luminescence energy, E_G and E_A the band-gap and acceptor energies, respectively, and T_e is the electron temperature. Figures 1(a) and 1(b) show the differential change in the e-A^0 line of GaAs at the electron CR when the microwave power (-15 dB) is modulated. We find that the electron effective temperature increases from ~8 K without microwaves to ~11.5 K.[9] Free exciton lines also show CR carrier heating since collisions are very efficient in transferring energy between free carriers and excitons.[10] This is evidenced in Fig. 1(c), 1(d) for CdTe: Electron CR is detected on the LO-phonon–assisted exciton recombination line (X-LO), whose line shape is[11] $\sim\sqrt{E}\exp(-E/kT_x)$, where T_x is the exciton effective temperature.

The two lines e-A^0 and X-LO are found to be rather insensitive to hole CR which occurs only at the highest microwave powers[12] [Fig. 2(c)]. However, there exist other luminescence lines which are more sensitive to the hole temperature T_h, such as those due to excitons bound on ionized donors D^+-X and free holes recombining on neutral donors D^0-h, the former because of D^+-X formation through the capture of a free hole by a neutral donor, the latter because of its intensity and line shape dependence on T_h. The lines at 1.5133 eV in GaAs and 1.5925 eV in CdTe are attributed to one or the other of these two processes, but at present we are not able to differentiate between the two unambiguously.[13] It suffices for our purpose to know that both are sensitive to T_h. The hole CR in CdTe measured on such a line is shown in Fig. 2(b). Experimentally, we find that

CR can also be observed on all other luminescence lines, such as exciton bound to neutral donors (D^0-X) [Fig. 2(a)] or acceptors (A^0-X), donor-acceptor pairs (D^0-A^0). All these lines appear to be sensitive to carrier temperature, either through line shape, capture probabilities, or impact ionization.

Classically, the momentum collision time τ can be deduced from a line shape analysis of the CR absorption curve. In the CR scheme developed here, the observed CR curve is not directly related to the absorbed power P, as it depends on the relationships between $T_{e,h}$ and P, and between the luminescence intensities and $T_{e,h}$. However, at small differential temperature changes these relations can be taken as linear functions and the signal is expected to have approximately the classical shape. The values for $\omega_c\tau$, the mobilities deduced from there and the various effective masses are listed in Table I. The mobilities are significantly higher than the best Hall mobilities ever measured in GaAs and CdTe samples. The reason is that, under light illumination, ionized shallow impurities are partially photoneutralized or screened,[18] thus reducing ionized impurity scattering. At such low temperatures, phonon scattering can be neglected and we expect the mobility to be limited by neutral-impurity scattering. The measured electron mobility in GaAs, $\mu_e = 1.9\times10^6$ cm^2 V^{-1} s^{-1}, is in good agreement with the calculated value.[4]

TABLE I. Comparison between parameters deduced from the optically and classically detected cyclotron resonance in GaAs and CdTe.

	m_i/m_0	$\omega\tau_i$	μ_i
p-type GaAs [100] [a]			
electron ($i=e$)	0.066 ± 0.001	32	1.9×10^6
	0.0665 ± 0.0002 [b]		
light hole ($i=lh$)	0.086 ± 0.010	1.4	6×10^4
	0.082 ± 0.004 [c]		
heavy hole ($i=hh$)	0.47 ± 0.01	7.2	6×10^4
	0.45 ± 0.02 [c]		
n-type CdTe [110] [d]			
electron ($i=e$)	0.096 ± 0.003	8	3.5×10^5
	0.0963 ± 0.0008 [e]		
light hole ($i=lh$)	0.12 ± 0.02	~4	~10^5
	0.102 [f]		
heavy hole ($i=hh$)	0.81 ± 0.05	9	4.5×10^4
	1.31 [f]		

[a] Hall measurements: $p_{300\,\mathrm{K}} = 1.3\times10^{14}$ cm^{-3}, $\mu_{h,\,77\,\mathrm{K}}$ (Hall) = 9500 cm^2 V^{-1} s^{-1}.

[b] Ref. 14.

[c] Ref. 15.

[d] Hall measurements: $n_{300\,\mathrm{K}} = 1.3\times10^{14}$ cm^{-3}, $\mu_{e,\,35\,\mathrm{K}}$ = 91 000 cm^2 V^{-1} s^{-1}.

[e] Ref. 16.

[f] Ref. 17.

Resonant carrier heating by CR absorption is a general phenomenon, allowing the technique to be applied to other materials. The microwave power requirements are low and can be met by standard equipment. From considerations regarding the field distribution in our cavity we can estimate the power absorbed per carrier to be ~10^{-13} W at resonance, resulting in an effective temperature increase ~2–3 K, in accordance to dc carrier heating measurements.[8] Similar temperature changes should be attained in other materials, as carrier relaxation rates are quite the same for various semiconductors.[19] As most luminescence lines are sensitive to such temperature changes, we therefore expect optical detection of CR to be feasible in a variety of materials. Moreover, the universal requirement of CR, namely $\omega_c\tau \gtrsim 1$, is more easily met because of the neutralization effect of the optically excited carriers.

We wish to thank H. L. Störmer for useful comments and Mr. Chamel for highly appreciated technical assistance. The GaAs samples were kindly supplied by K.-H. Zschauer (Siemens) and the CdTe samples by R. Triboulet (Meudon-Bellevue). The Laboratoire de Spectrometrie Physique is a Laboratoire associé au Centre National de la Recherche Scientifique. The Laboratoire de Physique de la Matière Condensée is a Groupe de Recherche du Centre National de la Recherche Scientifique.

[(a)] Present address: Bell Laboratories, Murray Hill, N. J. 07974.

[1] For a recent review, see C. R. Pidgeon, in *Handbook on Semiconductors*, edited by M. Balkanski (North-Holland, Amsterdam, 1980), Vol. 2, p. 223.

[2] For an historical account of the technique, see the review by B. Lax, J. Magn. Magn. Mater. 11, 1 (1979).

[3] C. Hermann and G. Lampel, Phys. Rev. Lett. 27, 373 (1971); S. Geschwind, in *Electron Paramagnetic Resonance*, edited by S. Geschwind (Plenum, New York, 1972), Chap. 5.

[4] G. E. Stillman and C. M. Wolfe, Thin Solid Films 31, 69 (1976).

[5] J. C. Hensel and K. Suzuki, Phys. Rev. B 9, 4219 (1974), and references therein.

[6] H. R. Trebin, U. Rössler, and R. Ranvaud, Phys. Rev. B 20, 686 (1979).

[7] R. Ulbrich, Phys. Rev. B 8, 5719 (1973).

[8]R. G. Ulbrich, Solid-State Electron. 21, 51 (1978).
[9]At such carrier temperatures and resonance frequency, $\hbar\omega_c \ll kT_e, kT_h$, several Landau levels are occupied, and quantum resonances in the valence band (due to the unequal spacing of the lowest Landau levels) are not observable. We only detect the classical hole resonance as shown by R. C. Fletcher, W. A. Yager, and F. R. Merritt, Phys. Rev. 100, 747 (1955).
[10]C. Weisbuch, Solid State Electron. 21, 179 (1978).
[11]E. F. Gross, S. A. Permogorov, and B. S. Razbirin, Fiz. Tverd. Tela (Leningrad) 8, 1483 (1966) [Sov. Phys. Solid State 8, 1180 (1966)].
[12]For completeness, we should mention that at such highpowers (0 dB, 200 mW) the electron CR in GaAs exhibits nontrivial thermal effects which are not yet fully understood.
[13]See R. Ulbrich and B. Moreth, Solid State Commun. 14, 331 (1974); D. Bimberg, W. Kottler, S. Suga, F. Willmann, and W. Dreybrodt, Verh. Dtsch. Phys. Ges. 9, 649 (1974), for GaAs; P. Hiesinger, S. Suga, F. Willmann, and W. Dreybrodt, Phys. Status Solidi (b) 67, 641 (1975); A. Nakamura and C. Weisbuch, Solid State Electron. 21, 1331 (1978), for CdTe. A discussion of the conditions under which one or the other of the two recombination processes is observed is given by P. J. Dean, Phys. Status Solidi (b) 98, 439 (1980).
[14]J. M. Chamberlain, P. E. Simmonds, R. A. Stradling, and C. C. Bradley, in *Proceedings of the Eleventh Conference on the Physics of Semiconductors, Warsaw, 1972*, edited by M. Miasek (PWN-Polish Scientific, Warsaw, 1972), p. 1016.
[15]M. S. Skolnick, A. K. Jain, R. Stradling, J. Leotin, J. C. Ousset, and S. Ashkenazy, J. Phys. C 9, 2809 (1976).
[16]A. Mears and R. Stradling, Solid State Commun. 7, 1267 (1967).
[17]Calculated band parameters from P. Lawaetz, Phys. Rev. B 4, 3460 (1971), injected in the expressions given in the appendix of Ref. 5.
[18]M. A. Paesler and H. J. Queisser, Solid State Commun. 21, 1143 (1977).
[19]E. M. Conwell, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1967), Suppl. 9.

Magnetic Fine Structure in Superconducting Tunneling: Spin-Glass Superconductors

M. J. Nass and K. Levin
The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

and

G. S. Grest
Department of Physics, Purdue University, West Lafayette, Indiana 47907
(Received 2 June 1980)

A superconducting spin-glass is studied as an example of how magnetic ordering is reflected in the superconducting density of states $N_s(\omega)$. Of particular interest is the prediction of a large "magnon" associated peak in $dN_s/d\omega$ for ω near the magnetic ordering temperature. This effect may have been observed in (gapless) superconducting spin-glasses. It is expected that this effect will also be present in antiferromagnetic superconductors.

PACS numbers: 74.50.+r, 74.70.Lp, 75.50.Kj

One of the greatest successes of the microscopic theory of superconductivity was the detailed understanding it yielded of superconducting tunneling experiments. Rowell and McMillan[1] showed how, by using the measured superconducting density of states $N_s(\omega)$, one can accurately extract an (averaged) phonon state density. In view of the recent excitement concerning intrinsic magnetic superconductors ($R\mathrm{Rh}_4\mathrm{B}_4$ and $R\mathrm{Mo}_6\mathrm{S}_8$, where R = rare-earth element) it is clearly of interest to look for *magnetic*-excitation–induced structure in $N_s(\omega)$. The hope is that one can extract detailed information about the dynamical form factor $S(q, \omega)$, in analogy with what was successfully done for the phonon case. As yet, no tunneling experiments in the coexistent phase have been done in these materials. There have, however, been successful measurements on an extrinsic (i.e., induced) superconducting spin-glass *Ag*Mn in proximity with Pb.[2]

Since the spin-glasses are also of considerable current interest and since tunneling measurements can help elucidate their magnetic properties, in this Letter we address these experiments. We predict features in the fine structure of $N_s(\omega)$ associated with the magnetic excitations, which may have been observed[3] experimentally. Our immediate goals are threefold:

(1) To set up the theoretical framework, and solve the resulting equations numerically, for computing $N_s(\omega)$ as a function of both frequency ω and T for magnetically ordered superconduc-