MAGNETOOSCILLATIONS OF THE ELECTRON–HOLE-DROP LUMINESCENCE IN GERMANIUM

K. BETZLER *, B.G. ZHURKIN, A.L. KARUZSKII and B.M. BALTER

P.N. Lebedev Physical Institute, Academy of Sciences of the USSR, Moscow, USSR

Measurements of the magnetoluminescence from electron–hole drops (EHD) were performed; intensity, halfwidth, and decay time of the EHD line show up oscillations as a function of the magnetic field in the region between 5 and 30 kOe. These oscillations can be explained by magnetooscillations of the carrier density inside the EHD due to oscillatory parts in the free energy of the electron–hole plasma.

At temperatures below 6 K and high enough excitation densities excitons in pure germanium condense into electron–hole drops (EHD) [1–3]. This condensed phase can be described as a quasimetallic plasma with a carrier density n_0 which is defined by the minimum of the free energy of the electron–hole system [4–6]. According to Keldysh and Silin [7], in a magnetic field the free energy of the EHD should exhibit a field-dependent oscillatory part. This should give rise to oscillations of n_0, as − in contrast to metals − this quantity is not fixed in the EHD by e.g. geometry conditions. Measurements of the EHD luminescence under the influence of a magnetic field for the first time were carried out by Bagaev et al. [8], who attributed the magnetooscillations they found in the integrated intensity of the EHD luminescence to oscillations of the binding energy for electrons and holes in the droplets. The influence of a magnetic field on the kinetics of the EHD line was studied by Gladkov et al. [9] who found a steady decrease of the EHD lifetime with increasing magnetic field. In the present paper we present extended time-dependent measurements of the magnetooscillations of the EHD luminescence. We can show for the first time, that − as proposed in ref. [7] − these oscillations can be referred to oscillations of the EHD density as a function of magnetic field. On the basis of a simple recombination kinetic model, conclusions on the quantum efficiency and the nonradiative processes in the EHD as well as on the difference between TA and LA-phonon assisted lines are possible.

The measurements were carried out on Ge samples with an impurity concentration of about 10^{12} cm^{-3} and a typical size of $5 \times 5 \times 0.3$ mm^3. They were immersed

---

* On leave from Physikalisches Institut der Universität Stuttgart; present address: Universität Osnabrück, FB 4, D 45 Osnabrück, Postfach 4469.

in liquid helium which was pumped to about 1.5 K. For carrier generation a pulsed GaAs laser was used with a peak power of 5 W, pulse length of 2 μs and duty factor of 0.2%. The power density on the sample was about 5 W cm^{-2} during the pulse. A magnetic field in Faraday configuration could be applied to the samples by means of a superconducting coil ($H < 32$ kOe) parallel to the (100) axis; this is the simplest case for Ge as for this configuration all conduction-band valleys are equivalent. The detection system consisted of an MDR-2 grating monochromator — here mainly used as a narrow band filter for the different EHD lines — and a Ge photodiode (risetime $\lesssim 1 \mu$s). For time-resolved detection, boxcar integration technique was used.

The time-dependent properties of the EHD can be described by a set of three coupled differential equations for the carriers inside the EHD, the excitons, and free electrons and holes, respectively. Under our experimental conditions of relatively high excitation and low temperature we can (for not too long times after excitation) neglect the influence of the exciton and free electron system on the EHD. This can be proved by the strongly exponential decay of the EHD luminescence under these experimental conditions [10,11]. So the time dependence of the mean carrier density in the sample after excitation can be described by one equation, only taking into account carriers inside the EHD:

$$d(Vn)/dt = -V(An + Bn^2 + Cn^3) , \qquad (1)$$

where V is the volume filled with droplets, n is the carrier density inside the EHD, and A, B and C are the recombination coefficients for impurity induced nonradiative, radiative, and Auger-like nonradiative processes, respectively. Higher order processes as for instance four-particle recombination processes (e.g. two-electron band-to-band transitions) can be omitted because of their extremely low recombination coefficients [12,13]. From eq. (1) quantum efficiency Q and lifetime τ can be defined; for the luminescence intensity we get

$$i(t) = \text{const } V(t) Bn^2 , \qquad (2)$$

after pulsed excitation, where $V(t) = V(0) \exp(-t/\tau)$. Integration leads to

$$I = \int_0^\infty i(t)\,dt = \text{const } V(0) n Q , \qquad (3)$$

with

$$Q = \frac{Bn}{A + Bn + Cn^2}, \qquad \tau^{-1} = A + Bn + Cn^2 . \qquad (4)$$

Fig. 1 shows the measured intensities $i(t)$ of the LA-phonon assisted EHD line (709 meV) as a function of the applied magnetic field. The intensities are normalized to their values at zero field. Oscillations with 2 maxima and 2 minima can be identified; the amplitude of the oscillations changes significantly with t, the time after excitation. This fact clearly shows, that the oscillations cannot be due to changing
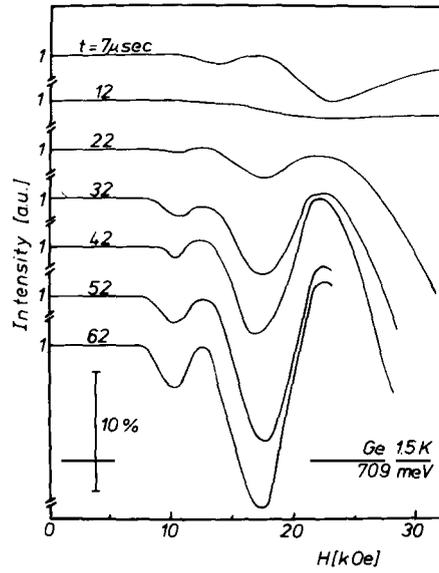
Fig. 1. Intensity oscillations of the LA-phonon-assisted EHD line for different times *t* between excitation and detection. All curves are normalized to the intensity at $H = 0$.
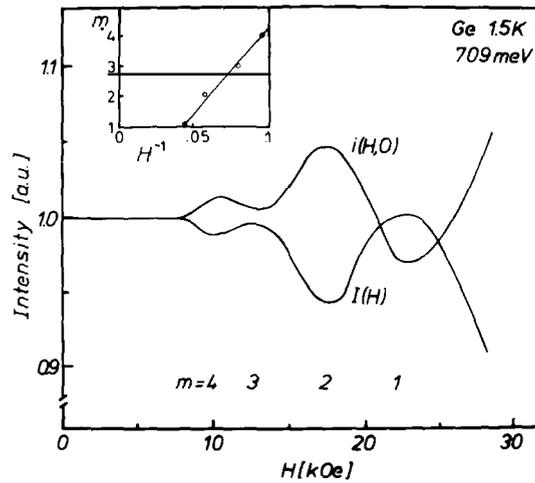

Fig. 2. Oscillations of the luminescence intensity at $t = 0$ and of the time-integrated luminescence intensity (calculated from the measurements in fig. 1). In the insert the maxima and minima positions are plotted as a function of the reciprocal field.

binding energy as suggested previously [8], but must be connected with varying τ. As A, B, and C should be constants at this relatively low fields, varying τ can only be caused by varying n (see eq. (4)). Assuming a magnetic field dependent variation $n(H) = n_0 + \Delta n(H)$, from (1)···(4) the following expressions can be derived for intensity oscillations:

$$\Delta i(H, t)/i_0(t) = [\Delta n(H)/n_0][1 - t(Bn_0 + 2Cn_0^2)], \tag{5}$$

$$\Delta I(H)/I_0 = [\Delta n(H)/n_0](A - Cn_0^2)/(A + Bn_0 + Cn_0^2). \tag{6}$$

To get numerical results, from the measured oscillations those of $i(t)$ for $t = 0$ and those of I were derived (fig. 2). The curve $i(H, 0)$ according to eq. (5) directly gives the oscillatory variations of the carrier density inside the EHD as a function of magnetic field. These variations are less than those calculated by Keldysh and Silin [7] by about one order of magnitude. A comparison of $i(H,0)$ and $I(H)$ leads, using (5) and (6), to the condition $A/Cn_0^2 < 0, 1$. That means, that in the EHD impurity induced nonradiative processes can be neglected compared to Auger recombination. Under the assumption $A \ll 0.1\,Cn_0^2$, we can derive the quantum efficiency in the EHD to be $Q \approx 25\%$ (for larger A: $Q < 25\%$). This value is less by a factor of 2 and 3, respectively, than former estimations by Benoit à la Guillaume et al. [14] (50%) and Pokrovskii et al. [15] (80%). It should be pointed out, however, that our derivation for the first time has to take into account the EHD luminescence only and need not make any assumptions about other lines.

In the insert of fig. 2 the positions of the oscillations' maxima and minima are plotted as a function of the reciprocal field. In coincidence with the condition [7] $m = \mu/h\omega + \text{const}$ for the index of maximum (minimum) — where μ is the Fermi energy and ω the cyclotron frequency of the electrons [16] — a linear dependence
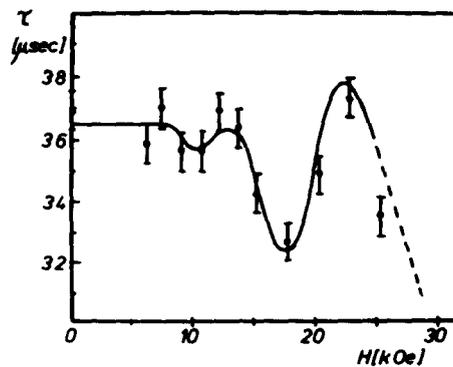

Fig. 3. Lifetimes of the EHD as a function of the magnetic field. The full line gives the calculated dependence.

is found. From the fitted μ, the carrier density inside the EHD can be derived to be 2.3×10^{17} cm^{-3} (band-structure parameters as in ref. [7]), which is in excellent agreement with values gained from the line shape [17].

As a further proof for the proposed model of magnetooscillations of the equilibrium density, EHD lifetime and line width were measured for different magnetic fields. Both show oscillatory behaviour; for the lifetime, the measured values, together with those calculated from the intensity oscillations (full line), are shown in fig. 3. For the calculation τ-oscillations were derived from the density oscillations using eq. (4).

For the TA-phonon-assisted EHD line (729 meV) a slightly different oscillatory behaviour was found which is shown in fig. 4. For a direct comparison the amplitudes of the last measured oscillations for LA and TA line respectively, are plotted as a function of delay time in fig. 5. Both show a rather good linear behaviour (eq. (5)), but there is some parallel shift which is connected with the fact that the TA-assisted radiative transition is not allowed at the band extrema in Ge [18]. For the intensities of the LA and TA line respectively, we can write:

$$I_{LA/TA} = \text{const}_{LA/TA} \int |M_{LA/TA}(k)|^2 D(k) d^3 k_e d^3 k_h , \qquad (7)$$

where $D(k)$ is the combined density of the occupied electron and hole states, and $M(k)$ is the total matrix element for the transition. For the LA line, M should be approximately constant in the region of integration, whereas for the rth order for-
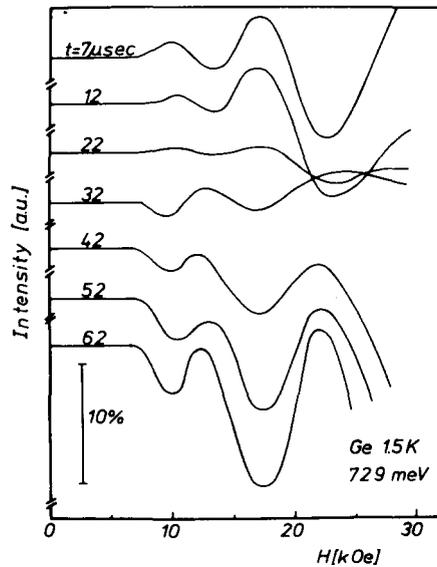

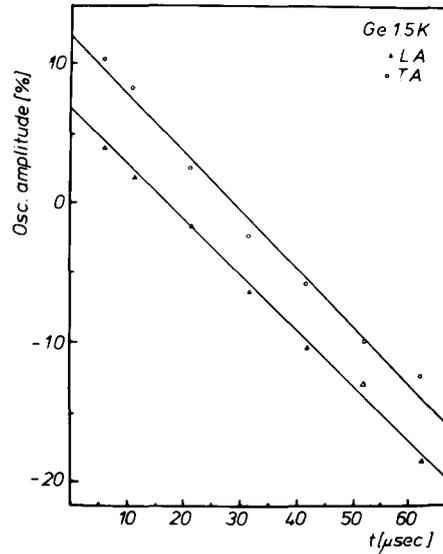Fig. 4. Magnetooscillations of the TA-phonon-assisted line.

Fig. 5. Plot of the amplitudes for the last measured oscillations taken from figs. 1 and 4 as a function of delay time t.

bidden TA transition $M_{TA} \propto (k_e + k_n)^r - k_e$ and k_h = momenta of the recombining electron and hole measured with respect to the band extrema. $\langle |k_e| + |k_n| \rangle \, \alpha n^{1/3}$ may be written for the mean value used in the integration, from (7) so we can derive:

$$I_{TA}/I_{LA} = \text{const } n^{2r/3} \,, \tag{8}$$

or for the oscillations:

$$\Delta i(H, t)/i_0(t)|_{TA} = \Delta i(H, t)/i_0(t)|_{LA} + \tfrac{2}{3} r \Delta n(H)/n_0 \,. \tag{9}$$

Taking the amplitude of the last oscillation plotted in fig. 5 as a measure for $\Delta i/i_0$, we get $r \approx 1$, which confirms that in the EHD too, the TA-phonon-assisted radiative tra transition is forbidden in the first order.

Acknowledgement

References

[1] L.V. Keldysh, Proc. 9th Int. Conf. Phys. Sem., Moscow (1968) p. 1303.
[2] Ya.E. Pokrovskii, Phys. Stat. Sol. (a) 11 (1972) 385 and references therein.

[3] M. Voos, Proc. 12th Int. Conf. Phys. Sem. Stuttgart (1974) p. 33, and references therein.

[4] W.F. Brinkman and T.M. Rice, Phys. Rev. B7 (1973) 1508.

[5] M. Combescot and P. Nozières, J. Phys. C5 (1972) 2369.

[6] P. Vashishta, S.G. Das and K.S. Singwi, Phys. Rev. Letters 33 (1974) 911, and references therein.

[7] L.V. Keldysh and A.P. Silin, Fiz. Tverd. Tela 15 (1973) 1532.

[8] V.S. Bagaev, T.I. Galkina, N.A. Penin, V.B. Stopachinskii and M.D. Churaeva, Pis'ma ZhETF 16 (1972) 120.

[9] V.B. Ginodman, P.S. Gladkov, B.G. Zhurkin, N.A. Kazakov and N.A. Penin, FIAN Preprint No. 90 (1974)..

[10] R.M. Westervelt, T.K. Lo, J.L. Staehli and C.D. Jeffries, Phys. Rev. Letters 32 (1974) 1051.

[11] K. Betzler, B.G. Zhurkin, A.L. Karuzskii and B.M. Balter, FIAN Preprint No. 71 (1975).

[12] W. Zeh, K. Betzler and R. Conradt, Solid State Commun. 14 (1974) 967.

[13] K. Betzler, T. Weller and R. Conradt, Phys. Rev. B6 (1972) 1394.

[14] C. Benoît à la Guillaume, M. Voos and F. Salvan, Phys. Rev. Letters 27 (1971) 1214.

[15] Ya.E. Pokrovskii and K.I. Svistunova, Fiz. Tekh. Poluprov. 4 (1970) 491.

[16] According to ref. [7] the main contribution to the oscillations should be caused by the electron system, whereas the contribution of the holes can be neglected. This can also be shown from the measurements in ref. [8] for different field directions.

[17] G.A. Thomas, T.M. Rice and J.C. Hensel, Phys. Rev. Letters 33 (1974) 219.

[18] M. Lax and J.J. Hopfield, Phys. Rev. 124 (1961) 115.

Discussion

J.C. Hensel: I notice that in your rate equation the Auger recombination term contains the density to the third power, indicative of a three-carrier Auger process. However, a three-carrier process is very unlikely without the assistance of a phonon in the e–h drop because of the lack of a suitable final state in the band structure to which the "odd" carrier can scatter. It would seem rather likely that the relevant process is a four-carrier Auger decay which is free from this restriction.

K. Betzler: You are right in pointing out that a three-carrier process without phonon-participation is very unlikely. We assume a process in which one phonon is emitted. Such a process have higher probability than a four-carrier process and was also found for higher temperatures in germanium. The phonon participation is included in the C-value, as it does not depend on the carrier density.

K. Morigaki: Did you make the measurement as a function of donor concentrations in As-doped germanium?

K. Betzler: No, we only measured in this medium donor concentration, because here the zero-phonon and the LA-phonon assisted lines have about the same intensities. The fact, that the two lines show different magneto-oscillatory behaviour may be explained by the model that the lines are caused by different states. This should also be the case at a changed donor-concentration, so that one should see a similar effect.

R. Conradt: I would make a comment on the question for a 4th order recombination: We have measured the radiative recombination of non-equilibrium Auger-carriers between E_g and $2E_g$ in Si. This spectrum was essentially flat indicating that single Auger-particles take up the whole energy. Therefrom we can conclude that the main impact recombination is the usual 3rd order Auger-recombination.