Valley-dependent spin polarization and long-lived electron spins in germanium

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Spin orientation and relaxation of conduction band electrons in bulk Ge are addressed by studying the steady-state circular polarization of the indirect gap photoluminescence (PL) at low temperatures. This provides a direct experimental proof of recently predicted spin-dependent selection rules for phonon-mediated optical transitions in Ge. In addition, we observe valley-dependent circularly polarized emission, and map the concomitant redistribution of electron spins within the multi-valley conduction band of Ge by gathering simultaneous access to the circular dichroism of light emitted across the direct and the indirect gap transitions. Finally, the lifetime of L-valley electrons is measured by means of decay curves of the indirect gap PL emission, yielding spin relaxation times in the order of hundreds of ns.

Ge offers a high light-matter interaction thanks to the close energy proximity between the fundamental conduction band (CB) minimum at the L point of the Brillouin zone and the relative minimum at the zone center Γ. This allows efficient coupling between the angular momentum of photons and the spin angular momentum of carriers providing spectroscopic access to its spin physics.[1,2] The spin-polarized electrons photogenerated at the Γ-valley of Ge are likely to experience scattering to the lateral side valleys at a rate that depends upon their excess energy.[3,4] In semiconductors like Ge and GaAs such intervalley relaxation mechanism has been shown to be responsible for a wealth of phenomena such as the intervalley spin transfer[5–7], the helicity-switching of direct gap emission[8], and the optically pumped spin Hall effects.[9,10] Hence the study of open issues in the context of the spin dynamics in the multi-valley CB of Ge is expected to further enrich the spin-dependent properties reported to date in nonmagnetic materials.

In this work we apply continuous-wave (CW) polarization-resolved photoluminescence to simultaneously address the circular dichroisms of light emitted across the direct and the indirect band gaps. This provides an experimental proof of the selection rules for the phonon-assisted optical transitions in unstrained bulk Ge and allows us to demonstrate valley-dependent spin polarization. Indeed, we show that the intervalley energy relaxation of CB electrons yields two simultaneous non-equilibrium spin populations with opposite polarizations at the center and at the border of the momentum-space. In order to complete the picture of the optical orientation process in Ge we then gather insight about the L-valley electron lifetime and its associated spin dynamics by means of PL decay time measurements of the indirect band gap emission.

To better capture the intrinsic spin lifetime of CB electrons we focus on a high purity bulk (001)Ge wafer, having a room temperature resistivity of 47 Ω cm. All the

measurements have been carried out below 100 K to ensure high spectral resolution and strong emission intensity. Optical spin injection was achieved by absorption of circularly $\sigma+$ polarized light through the direct gap and the excitation energy was fixed at 1.165 eV during both CW and pulsed PL measurements. For the former, we used the 1.165 eV line of a Nd:YAG laser at about 4 KW/cm^2 power density. The PL was detected by a dispersive system followed by an InGaAs photodiode array working in the 0.56 - 1.55 eV range. The state of PL polarization was studied by a polarimeter analyzer as discussed in Refs. 7 and 11. We recall here that the modulation of the PL intensity versus the analyzer angle θ provides the Stokes parameters and in turns the polarization type and degree (ρ) of each spectral line.[12] We carried out time-correlated single photon counting experiments using a Nd:YAG Q-switched laser, whose pulses have a temporal width of about 10 ns and energy between 39 μJ and 122 μJ. In this set-up the laser light was focused on a 500 μm diameter spot at the sample surface, and the emission was probed by a monochromator coupled to a photomultiplier tube (Hamamatsu R5509-73). The band pass was between 3.6 nm and 7 nm, and the time resolution of the whole detection system was ~2 ns.

Figure 1(a) shows the CW PL spectrum of the bulk Ge wafer at 4 K. The PL is dominated in the 0.70 - 0.75 eV range by peaks related to the transitions from the L-valley of the CB (cL) to the top of the valence band (VB) at the zone center Γ ($v\Gamma$). In particular, the three spectral lines at 704 meV , 712 meV, and 733 meV are respectively due to the transverse optical (TO), longitudinal (LA) and transverse (TA) acoustic phonon-assisted cL-$v\Gamma$ recombinations.[13] The shoulder at about 0.67 eV has been recently ascribed to the two-phonon mediated cL-$v\Gamma$ emission.[13] Finally, the weak PL feature at about 0.88 eV is due to the direct gap $c\Gamma$-$v\Gamma$ transition involving electrons and holes in the vicinity of the Γ point.[8,14]

Before addressing the quantitative investigation of the state of light polarization, we shall discuss the physics of the energy relaxation mechanisms as summarized in the inset of Fig. 1(b) and recently discussed in Ref. 8. Under the excitation conditions used in our experiments, elec-

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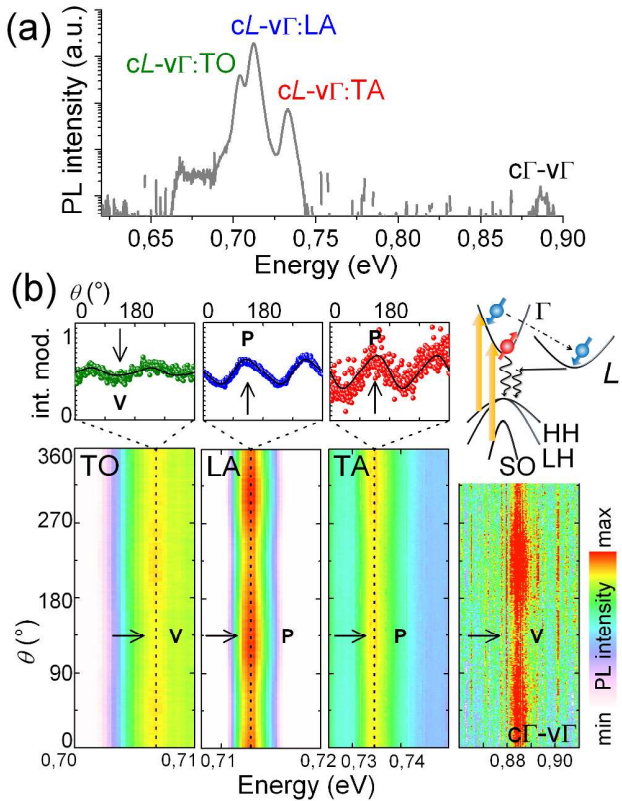b) fabio.pezzoli@unimib.it

FIG. 1. (a) PL spectrum of intrinsic bulk Ge at T = 4 K. (b) Lower panels: color-coded contour plots showing the amplitude of the cL-vΓ:TO, cL-vΓ:LA, cL-vΓ:TA, and cΓ-vΓ PL peaks. θ is the analizer angle. Peaks (P) and valleys (V) of the PL intensity are indicated at $\theta = 135°$. Upper panels: amplitude modulation of the indirect emission peaks normalized to their mean value, and sketch of the PL process in the sample following 1.165 eV excitation.

trons are photogenerated in the Γ-valley from optically coupled heavy hole (HH) or split-off (SO) VB states with opposite spin orientation. Noticeably the former provides electrons with a larger excess energy than the latter. In intrinsic bulk Ge, electron-phonon interactions drive energy relaxation of higher energy electrons via scattering towards X- and L-valleys.[3] As a result, the relatively larger contributions of the Γ-valley electron population excited from the SO has been shown to lead to a direct-gap luminescence having the opposite helicity as the one of the absorbed photons at the excitation energy.[8] By the same argument we do indeed expect that the vast majority of hot electrons excited from the HH are likely to be collected at the 4 equivalent L CB minima. For sufficiently long spin lifetimes this possibly results into transition across the indirect gap which shall be copolarized with respect to the laser excitation and counterpolarized with respect to the direct gap emission.

In the lower panels of Figure 1(b) we report color-coded maps showing the variation of the amplitude of the emission lines with the analyzer angle θ. The color scale evidences maxima or peaks (P) and minima or valleys (V). At first we note that all the four emission lines feature a sinusoidal behavior of the PL as a function of θ, which is the known fingerprint of circular polarization.[12] On one hand, this confirms recent observations made in bulk Ge of circularly polarized direct gap emission.[8] On the other hand, Fig. 1(b) discloses the PL circular dichroism of the indirect gap emission, finally demonstrating that the electron spins injected in the Γ states of bulk Ge withstand the intervalley Γ-to-L transfer and the long dwell time within the L-valleys. This is a remarkable result, since it has been so far expected that the long carrier lifetime would have hinder such an observation in an indirect bulk material. It should be noted that we can safely attribute the circular polarization to the nonequilibrium electron spin population created upon optical excitation, because the strong spin-orbit coupling in the bulk VB leads to a negligible contribution of holes due to their fast spin relaxation.[15]

In Fig. 1(b) the sinusoidal pattern of the PL intensity also indicates the helicity of the light polarization thus offering a deeper insight in the spin dynamics taking place in the multi-valley CB of Ge. We note that the amplitude modulation of the recombination across the direct gap, cΓ-vΓ, is shifted by 90° with respect to the one of the main indirect gap emission, cL-vΓ:LA (see the P and V values pointed out at $\theta = 135°$). This phase difference stems for cross polarized helicity. In particular, the map in Fig. 1(b) shows that the cΓ-vΓ line is *counter-circularly* polarized with respect to the $\sigma+$ excitation and, above all, it shows that the cL-vΓ:LA emission is *co-circularly* polarized with the $\sigma+$ excitation. Such findings are explained by the physical picture outlined above and provide a solid demonstration that by optical spin injection through the direct gap we are able to create two counter-polarized spin populations at the two CB minima, effectively locking the circularly polarized emission to the valley involved in the radiative recombination.

We shall now elaborate further on the role of phonons in dictating the polarization of the cL-vΓ transitions. In Si the spin-dependent selection rules were investigated both theoretically and experimentally in Refs. 16 and 17. Li et al. developed group theory analysis also for unstrained bulk Ge.[18] They provided the polarization degree ratios between the $\sigma+$ and $\sigma-$ components of the indirect gap phonon-assisted optical transitions, reporting helicity reversal between TO and the LA/TA contributions.[7,18] Our experimental data summarized in Fig. 1(b) show that both the cL-vΓ:LA and cL-vΓ:TA emission lines are indeed co-circularly polarized with respect to the excitation, whilst the cL-vΓ:TO transition is counterpolarized, despite its close energy proximity to the main LA peak. This finding fully matches with the theory and further corroborates the crucial role played by phonon symmetries in optical orientation experiments. The upper panels of Fig. 1(b) highlight the peak intensities as a function of θ of each of the indirect emission lines normalized to their mean values. It turns out that the
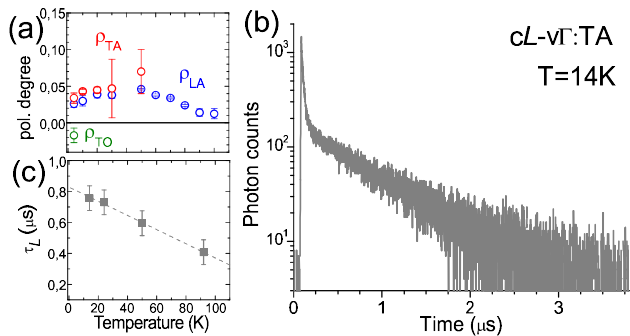
FIG. 2. (a) Temperature dependence of the polarization degrees of the phonon-assisted cL-vΓ PL peaks. (b) Decay curve for the cL-vΓ:TA emission measured under excitation with $7.6 \cdot 10^6$ W/cm^2 peak power density in the Ge sample at T = 14 K. (c) Temperature dependence of the slow component of the curve. The dashed line is a linear fit of the data.
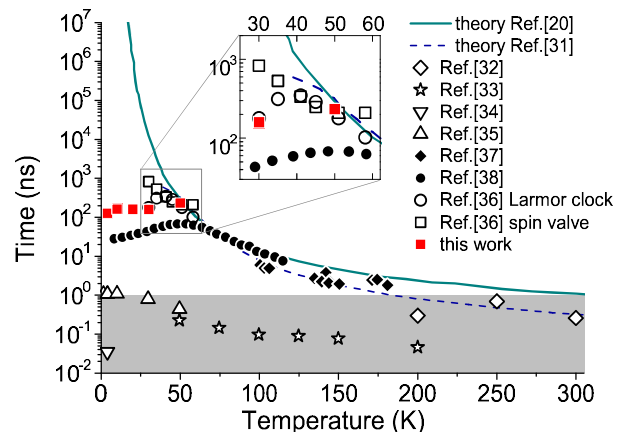

FIG. 3. Spin lifetime of CB electrons in bulk Ge as a function of the temperature. Red squares are data obtained in this work. Solid[20] and dashed[31] lines result from theoretical calculations. Empty symbols are values obtained by electrical measurements, reported in Ref. 32–36; full symbols are values obtained by optical measurements, reported in Ref. 37 and 38.

amplitude of the oscillations is very similar for all the PL features, which means similar polarization degrees. The results of the Stokes analysis[7] shown here as solid black lines superimposed to the raw data, provide the following polarization degrees: $|\rho_{LA}| = 2.6 \pm 0.5$ %, $|\rho_{TA}| = 3.4 \pm 0.7$ %, and $|\rho_{TO}| = 1.7 \pm 1.0$ %, where the subscripts label the phonons involved in the optical transitions.[19] We emphasize that these data compare well within the experimental errors with the phonon-dependent circular polarization properties identified by the theory, namely $|\rho_{TO}/\rho_{LA}| = 1$ and $|\rho_{TA}/\rho_{TO}| = 2$.[7,20]

Fig. 2(a) shows the temperature dependence of the polarization degrees of the phonon-assisted emissions from 4 K to 100 K.[21] ρ_{LA} presents a maximum at 50 K, while ρ_{TA} is larger than ρ_{LA} and reproduces its behavior. The following argument can be put forward to explain this finding: the decrease at higher temperatures can be viewed as the signature that the spin relaxation time (τ_s) becomes shorter than the carrier lifetime (τ_L), whereas the shallow reduction below 50 K might evidence the emergence of spin depolarization processes.

The quantitative comparison between the expected degrees of spin polarization and the measured circular polarizations for the phonon-assisted optical transitions allows us to further glimpse spin-loss mechanisms. Since the measured circular polarization degrees, albeit small, are non zero, we can infer that τ_s shall be comparable with τ_L, i.e. a fraction of the electrons spins are still oriented prior to radiative recombination across the indirect band gap. At a variance from analytical calculations that yields hour-long radiative lifetimes for a perfect bulk Ge[22], the experimental data of τ_L range from fractions of ns to several μs.[23,24] Since the actual lifetimes can be strongly dependent upon temperature, doping and crystal defects, we have studied the decay curve of the indirect PL emission to better estimate τ_L and the magnitude of τ_s in our sample.

Because of the detector cutoff at long wavelengths, from now on we focus among the phonon replicas on the higher energy cL-vΓ:TA line, which ensures us with a sufficient signal-to-noise ratio. The decay of its PL intensity, shown for T = 14 K in Fig. 2(b), presents two exponential components: a fast one, which vanishes in some tens of ns, and a slow one which lasts few μs. The former is ascribed to non-radiative Auger recombination, expected under pulsed excitation because of the initial high density of photogenerated carriers. From Fig. 2(b) we conclude that Auger processes affect the carrier lifetime at an early stage of the recombination dynamics. The decay time for this fast component is ∼22 ns, in good agreement with Auger recombination times previously reported in Refs. 23 and 25. Furthermore, when the pulsed excitation power is reduced, the weight of the Auger decreases until it becomes negligible [not shown]. In CW experiments this effect is also not significant due to the low excitation power density. The change of slope around 200 ns in Fig. 2(b) evidences the presence of a recombination process having a characteristic time of ∼757 ns that can be ascribed to the typical lifetime of L-valley electrons under low excitation conditions. It is likely that such slow component can be limited by competitive non-radiative channels due to the capture of carriers by bulk traps[26,27] and by surface states.[24] We finally point out that this value matches the range of τ_L extracted by low temperature electrical measurements.[28,29] Figure 2(c) reports the values of τ_L measured at temperatures from 14 K to 92 K. τ_L reduces when the temperature rises, as observed in other Ge-based systems[30], and it scales almost linearly in this temperature range.

In order to address the spin relaxation time for CB electrons we combine the measured τ_L and the circular polarization degree of the cL-vΓ:TA emission line [Fig. 2(a)]

as a function of the temperature.[1,39] By following the physical picture put forward above, the spin polarized population injected into L-valleys is made by pristine electrons excited from heavy and light holes, whose average contribution would lead by itself to an initial circular polarization degree $\rho_0 = 25\%$.[2,40] The theory regarding the spin-dependent phonon-assisted transitions can then be applied to extract the spin relaxation time from the measured circular polarization degrees, ρ_i, for each of the cL-vΓ peaks. We can thus write $\rho_i = (S_i \cdot \rho_0)/(1+\tau_L/\tau_s)$, where S_i results from the selection rules for the i-th phonon mode.[18] For the specific case of the cL-vΓ:TA line, this relation reduces to $\rho_{TA} = 0.25/(1 + \tau_L/\tau_s)$. It is worth noting that the spin relaxation time obtained by this simple approach provides a lower limit estimate since ρ_0 is not experimentally accessible.

Figure 3 displays the values of τ_s obtained in this work (red squares) together with a survey of recent theoretical[20,31] and experimental[32–38] values of the electron spin lifetime in Ge spanning the temperature range from 2 to 300 K. Empty (full) symbols are experimental data obtained by electrical (optical) methods, while the solid and the dashed line represent the intrinsic value of τ_s calculated in Ref. 20 and Ref. 31, respectively.

In Fig. 3 all the experimental data fall below or close to the theoretical curves. The latter refer to intrinsic relaxation mechanisms in perfect crystals, constituting an upper boundary for the experimentally accessible lifetime window. The area highlighted in gray in Fig. 3 demonstrates that electrical spin injection, mainly based on three terminal device geometries, provides values that are shorter than 1 ns over the whole temperature range. Above 200 K these data compare well with the theory, but below 200 K their discrepancy with the theory drastically diverges by several orders of magnitudes. Such observation is in line with the argument that at low temperatures a rapid spin relaxation channel can be provided by scattering with ionized impurities in the heavily doped region of the device[35] as well as with the crossover from spin accumulation into interface states to spin injection into the CB, recently identified around 200 K by Jain and coworkers.[32] Fig. 3 points out that the device fabrication as well as device-to-device fluctuations can significantly affect electrical spin injection and detection experiments, hampering the observation of spin lifetimes longer than few ns. In contrast to the electrical methods, all optical-based techniques do not need ferromagnetic contacts or high doping levels and thus are less prone to measurements artifacts. The same applies to transport data acquired under ballistic regime, which do not rely on spin accumulation at the interfaces.[36] Indeed as summarized in Fig. 3 above ∼50 K, τ_s is in the tens-of-ns range and the theory agrees well with the estimate of spin relaxation provided by magneto-optical measurements (full symbols)[37,38] and by hot electron devices (open circles and squares).[36] Below 50 K data obtained in this work (red squares) show hundreds-of-ns long spin relaxation times and better follows results obtained by Larmor clock

technique.[36] Noticeably in this temperature regime, a gap is opened between our data set and the ones by Faraday rotation experiments[38] and by spin valve devices under weak electric fields[36], despite the very similar doping level (few 10^{13} cm^{-3}) of the samples used in these three works. The reasons for such discrepancies might be possibly traced back to the actual differences in the measurement techniques, e.g. the presence of external fields and the different kinetic energies of the electrons probed by the experiments. Finally, our result increase by almost one order of magnitude the longest time previously reported below 30 K,[38] ruling out the role of the carrier lifetime τ_L as main limiting factor in the observed difference between the magneto-optical report of the spin lifetime T_s and the predicted spin relaxation times τ_s, being $1/T_s = 1/\tau_s + 1/\tau_L$. In the low temperature regime possible relaxation mechanisms such as exchange scattering between CB electrons and impurity carriers[41], hyperfine interaction[42] or impurity-assisted relaxation channels[43] may play a significant role. In addition, shake-up[44] and Coulomb scattering between electrons and holes[45] may be present in optical based experiments.

In conclusion, circularly polarized photoluminescence spectroscopy has been employed to demonstrate valley-dependent spin polarization in Ge and we explained this finding in terms of the energy relaxation of electrons via ultrafast intervalley scattering. Since in Ge the circular polarization degrees predicted for optical transitions mediated by the electron-phonon interaction[18,20] were still awaiting for experimental confirmation, we have tackle this issue by means of measurements of circular PL dichroism, providing a direct experimental proof of the spin-dependent selection rules in this material. Moreover, we employed time decay PL measurements of the phonon-assisted optical transitions to gather insight into spin relaxation mechanisms. We achieved spin relaxation times in the range of hundreds of ns at cryogenic temperatures and emphasized the presence of spin depolarization processes that are more efficient than the intravalley electron-phonon scattering. Finally, this work put forward the importance of optical studies aimed to deepen our understanding of spin injection in multi-valley semiconductors and emphasized the importance of systematic studies to reliably address the dominant processes dictating the spin relaxation in Ge in the low temperature regime.

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