Two-photon selective population of energy valleys in PbTe and jumps of the recombination-radiation polarization direction

I. P. Areshev, A. M. Danyshevskiĭ, S. F. Kochegarov, and V. K. Subashiev

A. F. Ioffe Physico-technical Institute, USSR Academy of Sciences
(Submitted October 16, 1976)
Pis'ma Zh. Eksp. Teor. Fiz. **24**, No. 11, 594–598 (5 December 1976)

We report observation of linearly polarized stimulated recombination radiation (RR) from a lead telluride crystal following two-photon pumping with linearly polarized radiation of wavelength 10.6 μ. The direction of the RR experiences a jump of ~90° when the direction of the polarization of the pump radiation (PR) (from −8 to +8°) in the crystal plane (100) undergoes a small change relative to the [010] direction. When the direction of the PR polarization is varied in the range −45–0°, the degree of polarization of the RR changes from 0.85 to 0.15. These facts are attributed to the onset of selective population of the energy valleys.

PACS numbers: 78.60.Dg, 78.20.Nv

We have previously reported[1] observation of stimulated recombination radiation (RR) from lead telluride crystals excited by radiation from a pulsed CO_2 laser ($\lambda = 10.6$ μ). The change of the direction of the linear polarization of the pump radiation causes an appreciable change (by up to 15 times) in the total RR yield. This effect was attributed to: 1) the strong dependence of the rate of excitation of the carriers in two-phonon absorption (TPA) in different valleys of the energy spectrum[1)] of the lead telluride on the pump polarization direction, 2) to the absence of intervalley transfer processes[2)] at sufficiently low temperatures, and 3) to the onset of stimulated radiation. In particular, it follows from expression (1) of[1] that in the case when the pump radiation propagates in a direction that coincides with the [100] axis, and the pump polarization direction makes an angle of 45° with the [010] axis, the rate of excitation into two valleys (e.g., [1, −1, 1] and [−1, 1, 1]) exceeds the rate of excitation into two other valleys ([1, 1, 1] and [−1, −1, 1]) by a factor of 6.25.

When the pump polarization plane is rotated 90°, the ratio of the carrier generation rates in the indicated valleys is reversed. On the other hand, when the polarization direction coincides with the [010] axis, the rate of excitation in all four valleys should be the same.

The presence of selective population of the energy valleys should lead to polarizability of the RR. The degree and direction of the polarization should depend substantially on the direction of the pump-radiation polarization.

The experiment aimed at observing this effect was performed on an n-PbTe sample ($n = 4\times10^{16}$ cm^{-3}) in the form of a freely mounted plate ~1 mm thick, in liquid He at 4.2 °K. The surfaces of the plate were dull after etching, and were not parallel. The beam of the pulsed CO_2 laser was directed along the [100] axis of the crystal. The laser radiation was linearly polarized. The radi-

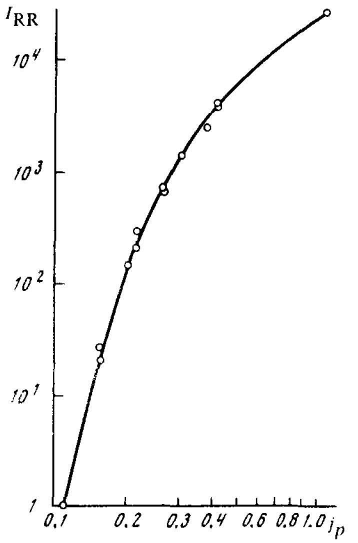

FIG. 1. Dependence of the recombination-radiation flux on the intensity of the pump radiation j_p. The maximum value of j_p corresponds to the absolute intensity $(7.5 \pm 5) \times 10^{25}$ quanta/cm^2 sec.

ation from the crystal was registered in the [100] direction from the front surface of the sample in a solid angle $\sim 22°$ ($\sim 4°$ in the crystal). The recombination radiation was focused and aimed directly on the photoreceiver (Ge: Au at 77 °K). We measured the dependence of the outgoing RR flux I_{RR} on the intensity j_p of the pump radiation (Fig. 1). At the smallest values of I_{RR} that could be registered, the $I_{RR}(j_p)$ plot turned out to be extremely steep ($I_{RR} \sim j_p^{20}$). Thus, even in the absence of a resonator, the RR gain in the crystal is very large and the observed emission is stimulated.

The next step was to place an analyzer (polyethylene diffraction grating) in front of the receiver and to measure the polarization characteristics of the RR. To check that there were no internal stresses in the crystal, we measured the polarizability of the RR under circular pumping. No linear polarization was observed in the RR. When a half-wave phase plate was placed in the laser beam, the radiation past the plate was linearly polarized and the polarization of the pump radiation was rotated.[3] In this case the RR was essentially linearly polarized. Figure 2 shows the obtained plot of the direction (ψ_m) and degree (ρ) of the linear polarization of the RR against the direction ϕ of the pump-radiation polarization. The angles ϕ and ψ_m are reckoned from the [010] direction in the (100) plane. It is seen from Fig. 2 that the direction of the RR polarization changes jumpwise from $\psi_m \approx -45°$ to $\psi_m = 45°$ when the angle ϕ changes from -8 to 8°. The degree of polarization of the RR decreases from $\rho = 0.85$ at $\phi = -45°$ to $\rho \approx 0.14$ at $\phi = -1°$.

It is easy to show that the degree and direction of the polarization of the RR should depend not only on the excitation direction but also on the RR observation direction relative to the crystal axes.

The connection between the RR signal I_{RR}, the direction ϕ of the pump polarization, and the position ψ of the analyzer ahead of the photoreceiver in the employed experimental geometry for spontaneous interband recombination excited by two-photon absorption of the pump radiation and in the absence of intervalley transitions, can be obtained in the form of the following expression:

$$I_{RR} \sim \left\{ \left[1 - \frac{\alpha}{3}(1+\sin 2\psi)\right]\left[1 - \frac{\alpha}{3}(1+\sin 2\phi)\right]^{2n} + \left[1 - \frac{\alpha}{3}(1-\sin 2\psi)\right] \times \left[1 - \frac{\alpha}{3}\left(1 - \sin 2\phi\right)\right]^{2n} \right\}, \qquad (1)$$

where $\alpha = (\gamma - 1)/\gamma$; $n = 1$ and 2 for linear and quadratic recombination, respectively.

It follows from (1) that when only one pair of valleys emits and the RR flux is measured in the [100] direction, the degree of polarization of the RR is $\rho = 0.437$.

In the experiment (Figs. 1 and 2) we observed stimulated emission with ρ reaching 0.85. This indicates that the polarization modes are appreciably enhanced in the crystal even in the absence of a resonator.

It follows also from (1) that in the case when the polarization direction of the pump radiation makes an angle of 45° with the [010] axis of the crystal ($\phi = -45°$ or 45° in Fig. 2) the direction of the RR polarization (ψ_m) should coincide with the pump-radiation polarization direction. From the data of Fig. 2 it is seen that this is indeed the case.

Attention is called to the fact that the 90° change in the direction of the RR polarization is not quite abrupt and that intermediate polarization directions ψ_m exist in a transition region of values of ϕ. If the radiation from the two

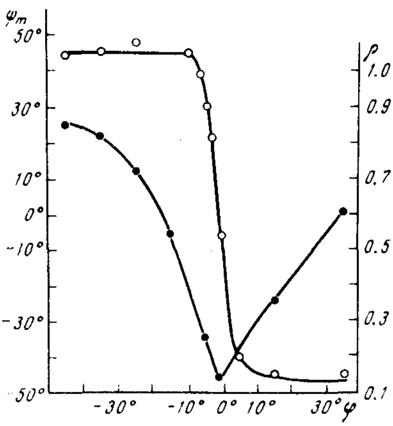

FIG. 2. Dependence of the direction and the degree of polarization of the recombination radiation on the direction of the polarization of the pump radiation. ϕ and ψ_m are the angles between the [010] of the crystal and the directions of the pump-radiation and recombination-radiation polarizations, respectively. The pump radiation is directed along the [100] axis of the crystal. ρ is the degree of polarization of the recombination radiation, light circles—$\psi_m(\phi)$, dark circles—$\rho(\phi)$.

pairs of valleys were to be independent, the direction of the RR polarization would be determined by the valley pair with the higher excitation rate at the given value of ϕ. In this case there could be only two states of polarization, $\psi_m = -45°$ and $\psi_m = 45°$. The presence of intermediate polarization directions can be explained in principle as being due to the small inclination of the sample plane to the (100) plane of the crystal. Another possible explanation is that inasmuch as the radiation of one pair of valleys stimulates partially recombination in another pair of valleys and vice versa, mutual phasing of the radiation is effected.

The authors thank E. L. Ivchenko for a useful discussion of the questions touched upon in the paper and L. S. Voronina for help with the experiment.

1) Lead telluride is a cubic crystal with a multivalley energy spectrum. The valleys are elongated along the [111] directions in the crystal. The degree of anisotropy of the reduced effective carrier mass, $\gamma = m_{\parallel}/m_{\perp}$, is ~11.4 according to the data of[2].

2) The minimal energy of the "intervalley phonons" in PbTe is ~3.2 meV.[3] The intervalley transfer due to the phonon scattering at low temperatures is therefore suppressed. On the other hand, intervalley transition on account of scattering by impurity centers and on account of "re-emission" processes[4] are not very effective in lead telluride, because of the large dielectric constant ($\epsilon = 400$) and the very low ionization of the shallow impurity levels.

3) Rotation of the phase plate did not change the pump intensity by more than 1.0%.

1. I. P. Areshev, A. M. Danishevskiĭ, S. F. Kochegarov, and B. K. Subashiev, Pis'ma Zh. Eksp. Teor. Fiz. **22**, 437 (1975) [JETP Lett. **22**, 211 (1975)].
2. Yu. I. Ravich, B. A. Efimova, and I. A. Smirnov, Metody issledovaniya poluprovodnikov v primenenii k khal'kogenidam svintsa PbTe, PbSe, PbS (Semiconductor Investigation Methods as Applied to the Lead Chalcogenides PbTe, PbSe, and PbS), Nauka, 1968.
3. G. W. Pratt, Jr. *et al.*, Proc. Tenth Intern. Conf. on Physics of Semiconductors, Cambridge, Mass., 1970, p. 87.
4. G. Weinreich, T. M. Wanders, Jr., and H. G. White, Phys. Rev. **114**, 33 (1959).