Submillimeter-Laser Magnetospectroscopy in Tellurium Making Use of the Nernst Effect

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Abstract—**In contrast to magnetotransmission and magnetophotoconductivity measurements in tellurium the radiation-induced Nernst effect reveals impurity and cyclotron transitions exactly and is not obscured by other effects as demonstrated in samples of different dislocation densities.**

I. INTRODUCTION

THERE has been a considerable effort on investigations of the submillimeter-laser magnetotransmission spectra of tellurium for both favorite orientations of the magnetic field direction relative to the symmetry axis of the crystal [1]. The transmission spectra of impure or deformed samples, however, are often obscured by extraneous effects and do not represent the real absorption structure. Multiple reflections at the sample surfaces, as well as the interference of the different normal modes, produce structures in the transmission spectra which must not be taken as typical for the absorption process [2]. Plastical deformation may produce dislocation inhomogeneities in tellurium, so that the usual condition of an optically uniform medium holds no longer for the interpretation of transmission data. To overcome these difficulties we measured directly the absorbed radiation energy in the sample independent of the specific boundary conditions of the electromagnetic wave. We applied a special cross-modulation technique using the Nernst effect and compared the results with the transmission spectrum of the same sample.

In Section II we give a detailed description of the experimental setup. The experimental results of transmission and different cross-modulation experiments for undeformed tellurium are discussed in Section III. Section IV presents the results for plastically deformed material. The summary is given in Section V.

II. EXPERIMENTAL ARRANGEMENT

Fig. 1 represents the schematic of the experimental arrangement. The submillimeter radiation is produced by a conventional HCN laser, which is pumped by an electric gas discharge. As laser fuel we used acetonitrile and ammonia or methane and nitrogen. The laser was tuned to a $\lambda = 337$-μm or $\lambda = 311$-μm wavelength by translation of the concave mirror. The radiation is coupled

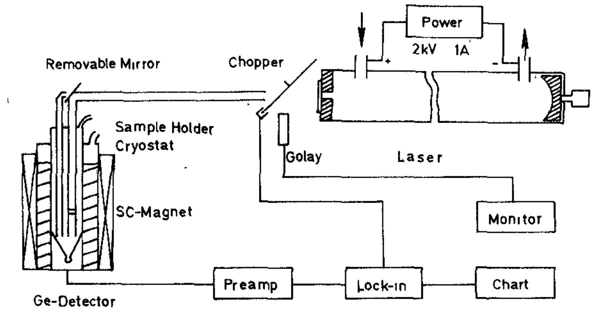

Fig. 1. Schematic of the experimental arrangement.

out of the cavity by a small hole in the plane mirror. Before entering the light-pipe system, the radiation is modulated by a 33-Hz chopper. The radiation intensity reflected from the chopper wings is detected by a Golay cell monitoring the laser output. The sampleholder is constructed as a separate cryostat in the 55-mm-ID bore of a 105-kG superconducting magnet. The Ge bolometer detecting the transmitted radiation intensity is integrated in the sampleholder. The influence of the strong stray field on the sensitivity of the Ge bolometer is exactly taken into account. For this calibration we used a separate light pipe bypassing the sample. In a transmission experiment the detector signal is processed by a conventional lock-in technique. Without any polarizer in front of the sample, the incident radiation superposes linearly polarized and unpolarized electromagnetic waves.

The investigated tellurium samples originated in the Czochralski grown single crystals HW Te 42 and HW Te 25 with a hole concentration of 3×10^{13} and 2×10^{14} cm^{-3}, respectively, at He temperature. The samples were cut by an acid saw [3] and chemically polished in Honeywell [4] and modified Honeywell [5] solution. The plane dimensions of the samples were about 4×6 mm^2. The thickness of the samples varied from 0.1 to 1.3 mm. The c axis was oriented perpendicular to the large sample surface and parallel to the magnetic field direction. The samples were provided with electrical contacts in a plane perpendicular to the field direction suitable for measurement of the Hall voltage and the transverse voltage drop across the sample. This voltage is composed of an unmodulated part $U_{2/4}$ and a cross-modulated part $\hat{U}_{2/4}$ which is modulated by the absorbed submillimeter radiation. In cross-modulation experiments this modulated part $\hat{U}_{2/4}$ was fed to the lock-in system instead of the bolometer signal. Even for zero current in the sample there are non-

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vanishing values of $U_{2/4}$ and $\hat{U}_{2/4}$ due to thermoelectric and thermomagnetic effects.

III. THE TRANSMISSION AND CROSS-MODULATION SPECTRA FOR UNDEFORMED TELLURIUM SAMPLES

Fig. 2 represents in the upper part the relative transmission and cross-modulation spectra of a pure and undeformed tellurium sample as a function of the magnetic field intensity for 337-μm wavelength radiation at a temperature of 2 K. The c axis of this sample with a hole concentration of 3×10^{13} cm^{-3} is parallel to the field direction. In the relative transmission spectrum we observe only one pronounced minimum due to the cyclotron resonance of free holes. There are no additional lines as observed in less pure material and caused by impurity transitions.

For zero current we observe a cross-modulation signal $\hat{U}_{2/4}$ having a typical negative phase shift relative to the modulation of the incident radiation. The origin of this signal is the radiation induced Nernst effect as proved later on. There is a complete correspondence of the extrema in the cross-modulation and transmission spectrum, which excludes any ambiguity in the interpretation. Hall-voltage and transverse magnetoresistance indicate no irregularities as seen from the lower part of Fig. 2.

In Fig. 3 we have plotted the corresponding data for a less pure tellurium sample with a hole concentration of 2×10^{14} cm^{-3}. In the relative transmission spectrum we observe in addition to the cyclotron resonance line several low- and high-field minima. These lines are produced by

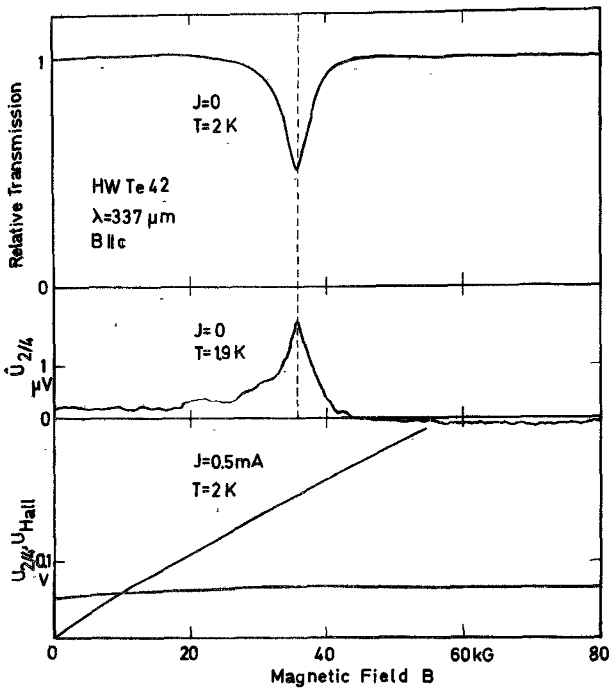

Fig. 2. The submillimeter-radiation properties of low-concentration tellurium as function of the magnetic field intensity demonstrating the correspondence of transmission and radiation-induced Nernst effect.

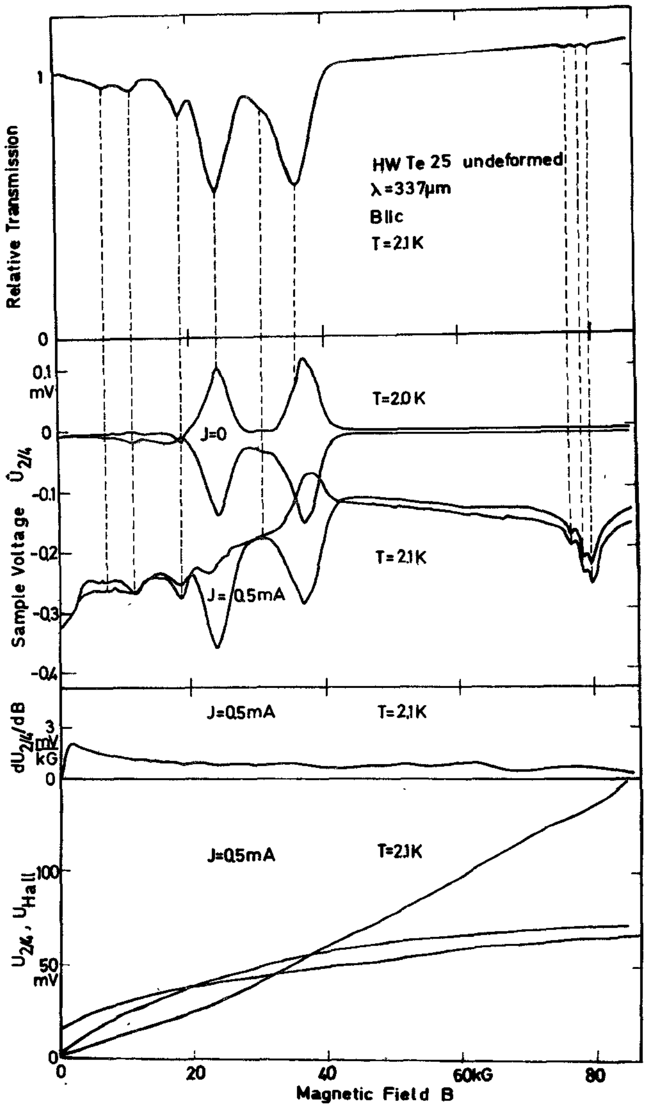

Fig. 3. The submillimeter-radiation properties of impure tellurium as function of the magnetic field intensity demonstrating the influence of impurity transitions on the different spectra.

transitions from impurity states, which are populated only at low temperatures. As demonstrated by the two relative transmission spectra in Fig. 4, the impurity lines disappear at higher temperatures and the cyclotron resonance increases in intensity. The high field triplet in Fig. 3 at about 80 kG has already been observed by Dreybrodt *et al.* [6] and Yoshizaki and Tanaka [7]. These authors, however, could not resolve the triplet structure. In the spectra of the cross-modulation signal $\hat{U}_{2/4}$ for zero and nonzero current and opposite magnetic field orientation in the middle part of Fig. 3 we observe again a correspondence of the extrema in comparison with the relative transmission spectrum.

In Fig. 5 we have plotted the complete set of cross-modulation spectra for different current values and opposite magnetic field orientations. The signal for zero current reverses sign for opposite field directions. There is a typical phase shift of about $-30°$ for this signal relative to the incident radiation. Because of both the sign reverse and the phase shift we explain the cross-modulation spectrum for zero current as produced by a thermo-

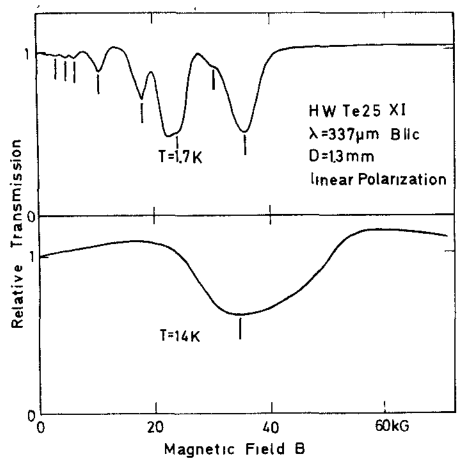

Fig. 4. The relative transmission spectra of impure tellurium for two different temperatures demonstrating the thermal freeze-out of impurity states at low temperatures.

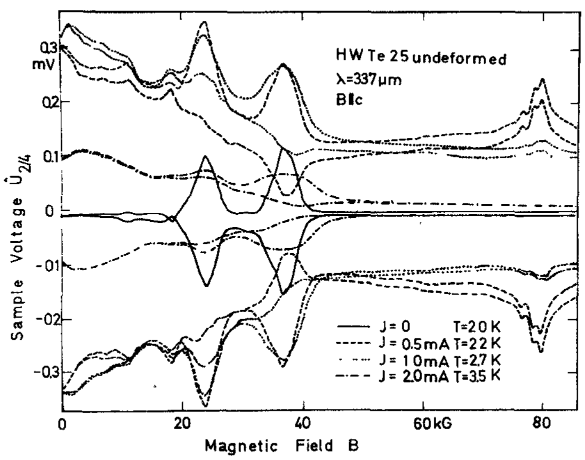

Fig. 5. The radiation induced cross-modulation signal versus magnetic field intensity indicating the different physical mechanisms for $J = 0$ and $J \neq 0$.

magnetic effect, namely, in this case, by the Nernst effect. The amount of the phase shift is determined by the heat capacity of the sample and the thermal contact to the surroundings. Due to the asymmetric thermal coupling of the sample by the five electric contact leads, the absorbed energy in the sample generates a transverse temperature gradient. This gradient in turn generates by the Nernst effect the cross-modulation signal $\hat{U}_{2/4}$, which is directly proportional to the absorbed radiation energy in the sample.

For nonzero current there is a superposition of thermomagnetic and photo effects exhibiting less pronounced structures. In the absence of an external magnetic field the modulated signal $\hat{U}_{2/4}$ has no phase shift relative to the incident radiation and depends in sign on the current polarity. This corroborates the interpretation as photo effect. In the strongly structured range of the spectrum between 20 and 40 kG we observe a magnetic field dependent phase shift of the signal. Because of this magnetic field dependence we cannot separate exactly the signal in photo and thermomagnetic parts by a simple addition or subtraction of the curves for opposite current and field polarity. With increasing current values the cross-modulation spectra become less pronounced and the peaks smear out. We attribute this effect to increasing temperature in the sample, which is even higher than the value measured at the sample surface.

Contrary to the spectra for zero current we observe for nonzero current a pronounced triplet structure corresponding to the triplet in the relative transmission spectrum. Vanishing phase shift of the signal relative to the incident radiation as well as the independence of field polarity indicates the photo effect as the operating mechanism. Because of the small line width we interpret the triplet as internal impurity transitions with subsequent excitation into the valence band [8]. This means that in our tellurium samples in high magnetic field intensities there exist impurities with a binding energy greater than 3.67 meV.

The cross-modulation signal for nonzero current can be affected by extraneous effects as demonstrated in Fig. 6. This sample originated in the same ingot HW Te 25 as the sample of Fig. 3. Despite the identical preparation technique, the cross-modulation signal is completely

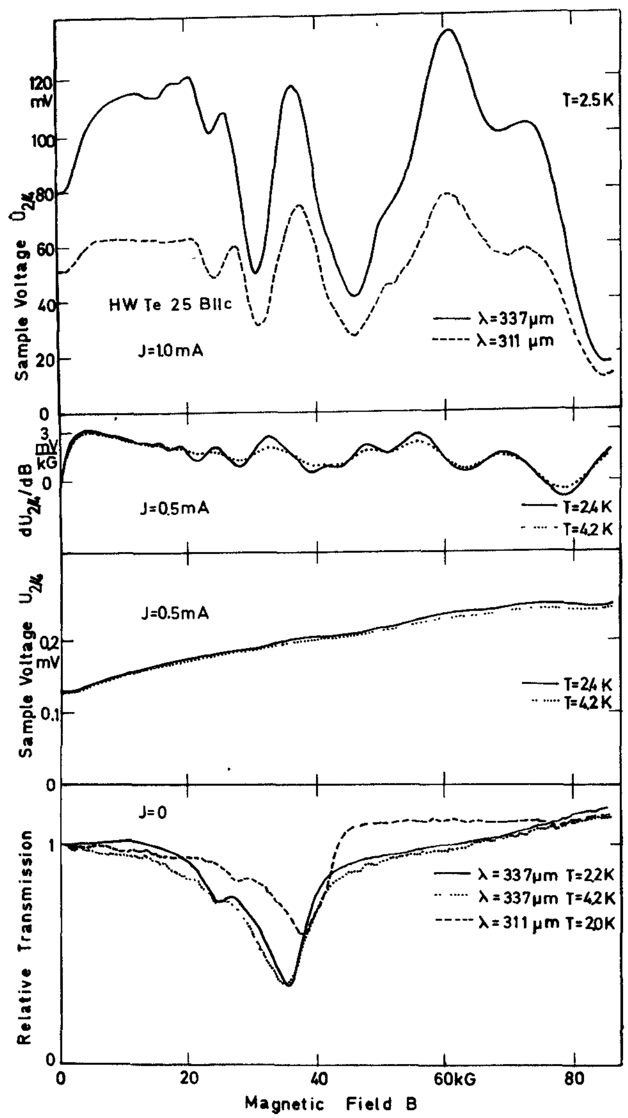

Fig. 6. The submillimeter-radiation properties of a tellurium sample completely determined by surface subbands as function of the magnetic field intensity.

determined by a thermomodulation of surface subbands. This can be seen from comparison of the relative transmission spectra and the oscillations in the transverse magnetoresistance in the middle part of Fig. 6. The signal of the cross modulation is phase shifted relative to the signal of the incident radiation. The spectrum is independent of the submillimeter wavelength as demonstrated by the application of 337 and 311-μm wavelength radiation. We explain the cross-modulation spectra as a thermomodulation of surface-subband oscillations in the magnetoresistance. These oscillations are very sensitive to temperature changes as shown by the two curves for $T = 2.4$ K and $T = 4.2$ K in the middle part of Fig. 6. Contrary to the cross-modulation spectrum, the relative transmission in the lower part of Fig. 6 exhibits the frequency dependent structure of the cyclotron resonance and impurity transitions. There seems to be no indication of the surface subbands characterized by an effective mass of $m = 0.14m_0$ [9].

IV. THE TRANSMISSION AND CROSS-MODULATION SPECTRA FOR DEFORMED TELLURIUM SAMPLES

The cyclotron resonance of plastically deformed tellurium with various dislocation densities has already been investigated [10]. In these experiments, however, the data of deformed and undeformed material originated from different sample individuals. To overcome this shortcoming we tried to investigate one and the same sample before and after plastical deformation. We deformed the tellurium sample of Fig. 3 without disconnecting the electrical contacts in the configuration for simplex-a-dislocations [11]. Because of the unfavorable ratio of sample thickness to sample width and the short-time application of the deformation force, we do not expect our results to agree with former investigations [10]. From the study of dislocation etch pits we have estimated an effective dislocation density of about 10^9 and 10^8 cm^{-2} for sample HW Te 25 A and B, respectively. In Fig. 7 we have plotted the relative transmission spectra of 337-μm radiation for the samples before and after plastical deformation. We notice a very large decrease in transmission after deformation. The transmission of sample HW Te 25 A was below the sensitivity of the detector. Even after a thickness reduction by a factor of 4.5 the transmission for zero magnetic field was only 0.5 percent of the value for undeformed material for this sample. Supposing that the transmission depends exponentially on the sample thickness, the transmission of the undeformed sample HW Te 25 A has decreased by a factor of 10^{11} after deformation! The less deformed sample HW Te 25 B exhibits a reduction in transmission by a factor of 50 for zero magnetic field. The structure of the relative transmission spectrum is completely different for deformed and undeformed material. We observe in the transmission spectrum of the deformed samples a broad minimum at a magnetic field intensity below the cyclotron resonance field. For magnetic field intensities higher than the cyclotron resonance field there is a strong increase in transmission. The impurity lines and cyclotron resonance line have nearly completely disappeared. Without any additional information the usual interpretation of the transmission spectra would result in a completely different absorption mechanism for deformed and undeformed material.

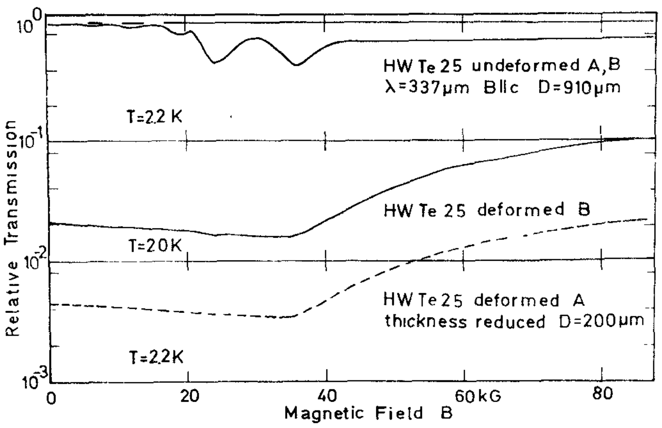

Fig. 7. The transmission versus the magnetic field intensity for undeformed and deformed tellurium samples demonstrating the tremendous decrease in transmission for deformed material.

The spectrum of the cross-modulation signal for zero current, that is, the spectrum of the radiation-induced Nernst effect, reveals, however, that the absorption of the radiation energy in the sample has not changed significantly as shown in Fig. 8. Especially the cyclotron-reso-

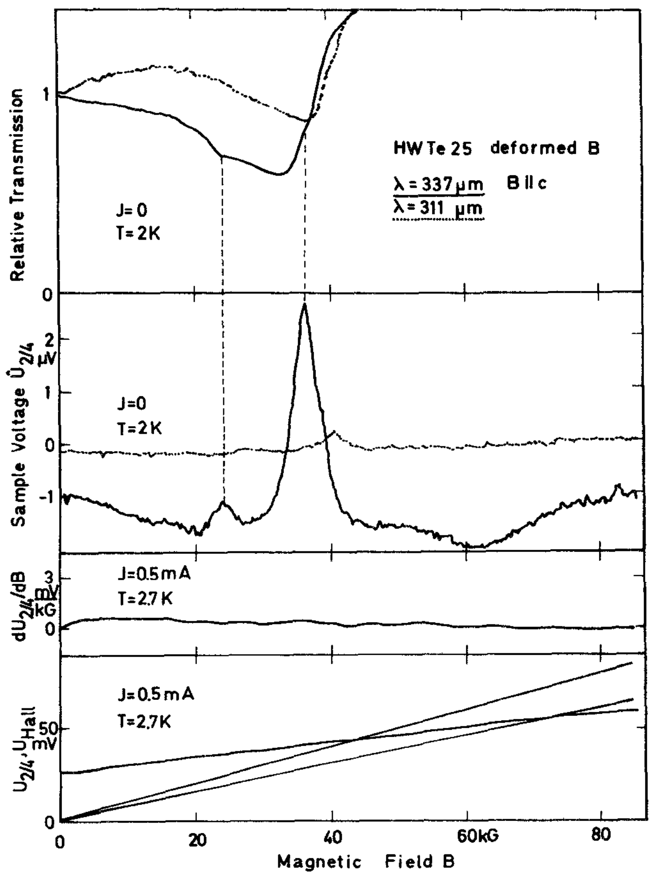

Fig. 8. The submillimeter-radiation properties of a deformed tellurium sample as function of the magnetic field intensity showing that only the transmission is affected by the deformation but not the cross-modulation signal of the radiation-induced Nernst effect.

nance line has the same field position and half-width as before deformation. The intensity of the impurity lines, however, has considerably decreased. We interpret this phenomenon not as an increase of the cyclotron-resonance intensity because the Hall voltage has decreased only by about 20 percent after deformation. This means that the concentration of impurity states has been considerably decreased by the deformation. The evident discrepancy of the relative transmission and cross-modulation spectrum is probably caused by dielectric inhomogeneities along the glide planes in the crystal. Therefore the deformed samples no longer represent an optical homogeneous medium. This however is a necessary condition for the usual interpretation of transmission spectra.

Fig. 9 demonstrates again the special advantage of the radiation-induced Nernst effect for the investigation of submillimeter-energy absorption in deformed tellurium. Whereas the zero current cross modulation represents the detailed absorption structure, the cross modulation for nonzero current is completely determined by other effects and provides no information about the magnetoabsorption process.

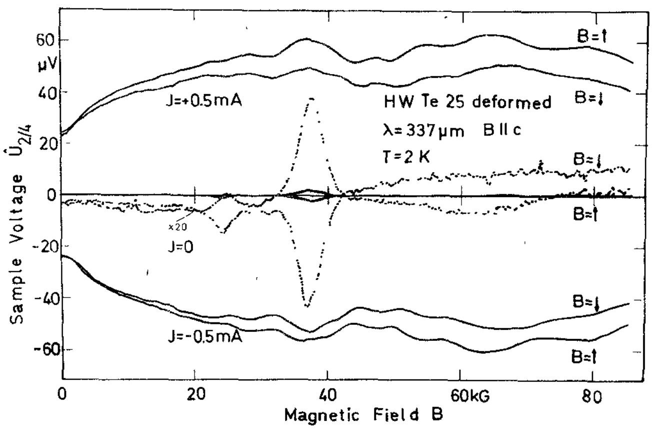

Fig. 9. The cross-modulation signal versus magnetic field intensity for deformed material indicating again the different mechanisms for $J = 0$ and $J \neq 0$.

V. CONCLUSION

We have demonstrated that both transmission and nonzero-current cross-modulation spectra can be strongly affected by extraneous effects and give no information about the magnetoabsorption process of submillimeter radiation in semiconductors. Contrariwise, the zero-current cross-modulation spectrum of the radiation-induced Nernst effect is directly proportional to the absorption of the radiation energy in the sample and provides a useful method to investigate the magnetoabsorption.

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