Definite Evidence for Population Inversion of Hot Electrons in Silver Halides

Susumu Komiyama and Taizo Masumi
Department of Pure and Applied Sciences, University of Tokyo, Komaba, Tokyo 153, Japan

and

Koji Kajita
Department of Physics, University of Tokyo, Hongo, Tokyo 113, Japan
(Received 5 August 1978)

Hot-electron kinetics and distribution in pure AgCl and AgBr at crossed electric and magnetic fields have been studied at 4.2 K up to $E_x = 3.4$ kV/cm and $H_z = 58$ kOe by detecting a response current Q_z to a perturbing electric field E_z in the transient condition. The results of measurements on $Q_z(E_x, H_z)$ established the pictures of an ideal streaming motion of electrons in the range $cE_x/H_z > V_{LO}$ (where $\frac{1}{2}m^*V_{LO}^2 \equiv \hbar\omega_{LO}$) and of *a population inversion of hot electrons* in the range $cE_x/H_z < V_{LO}$.

An anomalous distribution of hot electrons as predicted by Maeda and Kurosawa[1] has been observed experimentally in silver halides at 4.2 K in crossed electric and magnetic fields.[2] Here we report a definite evidence for the population-inversion phenomena obtained by successfully detecting an anomalous current flowing within a limited area K in velocity space.

The experiment was performed on photoelectrons in zone-refined AgCl and AgBr crystals, which have the values of low-field electron mobility about 3.6×10^4 and 1.2×10^5 cm^2/V sec, respectively, at 4.2 K. High electric fields $(E_x, 0, 0)$ up to 3.4 kV/cm and crossed magnetic fields $(0, 0, H_z)$ up to 58 kOe were applied to a specimen of a typical size about $0.7\times5\times5$ mm^3. A weak electric field E_z was also applied along the z direction and the response current Q_z was detected by a fast-pulse technique with an improved arrangement of blocking electrodes. Photoelectrons of the order of 10^7/cm^3 were excited in the specimen by using a xenon flash tube. No appreciable space-charge effects were involved because of the low concentration of photocarriers. The measurements were made in the transient condition.[3]

The response current Q_z was studied as a function of E_x and H_z. Dependence of Q_z on H_z at various levels of E_x is shown in Fig. 1 for a crystal of AgCl. In the measurement, E_z is fixed to 15 V/cm, which is sufficiently low so that the application of E_z does not cause any appreciable hot-electron effects by itself. Therefore we can regard Q_z as a probe current to the hot-electron system in the presence of E_x and H_z. The current Q_z at $E_x = 0$ is almost independent of H_z. This is naturally expected for AgCl, which has a spherical conduction bant at point Γ.[4] With increasing E_x, the current Q_z exhibits an overall decrease in a low-H_z range, reflecting a reduction in the electron mobility with E_x.[2] Every Q_z-H_z curve at high E_x includes a flat portion in a relatively low-H_z range, while it rises steeply with H_z in a higher-H_z range causing a kink at an intermediate H_z. The position of the kink shifts toward higher magnetic field as E_x increases. It was found that the following equation is satisfied at the kink:

$$\frac{cE_x}{H_z} = V_{LO}, \tag{1}$$

where c is the light velocity and V_{LO} represents the velocity of an electron whose kinetic energy is equal to the LO-phonon (LO stands for a longitudinal optical branch) energy $\hbar\omega_{LO}$ $(\frac{1}{2}m^*V_{LO}^2 \equiv \hbar\omega_{LO})$. The quantity V_{LO} is evaluated for elec-

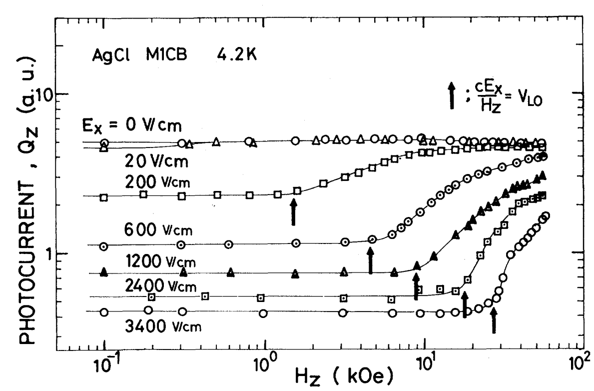

FIG. 1. Photocurrent Q_z in the z direction at crossed fields $\vec{E} = (E_x, 0, E_z)$ and $\vec{H} = (0, 0, H_z)$. Here the perturbing fields E_z (15 V/cm) is sufficiently low so that Q_z directly reflects the "electron mobility" along the z direction in the condition of $\vec{E} = (E_x, 0, 0)$ and $\vec{H} = (0, 0, H_z)$. An arrow on each Q_z-H_z curve indicates the magnetic field position at which $cE_x/H_z = V_{LO}$ (where $\frac{1}{2}m^*V_{LO}^2 \equiv \hbar\omega_{LO}$) is fulfilled. All the curves at high E_x rise steeply just above the indicated field.

trons in AgCl to be 1.4×10^7 cm/sec by using the value of the polaron effective mass[5] $m^* = 0.43m_e$ (m_e is the free-electron mass) and the value of $\hbar\omega_{LO} = 23$ meV.[6] The position of magnetic field at which Eq. (1) is fulfilled is indicated on each curve in Fig. 1 by an arrow. We can summarize the behavior of $Q_z(E_x, H_z)$ as follows: (A) In the range of relatively low H_z (with relatively high E_x) where $cE_x/H_z > V_{LO}$, Q_z is almost independent of H_z while it decreases with increasing E_x. (B) In the range of relatively high H_z (with relatively low E_x) where $cE_x/H_z < V_{LO}$, Q_z increases abruptly with H_z. Essentially similar results were obtained also for AgBr.

Let us try to interpret these results in terms of the streaming motion of electrons.[7,8] It has been confirmed in the Hall-effect measurements[2] and also in cyclotron-resonance experiments[9-11] that an electron in silver halides performs an ideal streaming motion at high electric fields in velocity space. A trajectory of an ideally streaming electron is shown by a solid arc in Fig. 2 for two cases of (a) $cE_x/H_z > V_{LO}$ and (b) $cE_x/H_z < V_{LO}$. When E_z is applied on a streaming electron, the time of free acceleration of the electron along the z direction is limited by successive emissions of LO phonon. The time interval between the successive emissions is equal to a traveling time of the electron from the point $|\vec{v}| = 0$ to a point $|\vec{v}| = V_{LO}$ on the arc. Therefore, in order to explain the observed increase of Q_z in the range $cE_x/H_z < V_{LO}$ in terms of the streaming motion only, we must consider that the traveling time increases abruptly in the range $cE_x/H_z < V_{LO}$. However, this is not the case. It can be easily shown that application of H_z (at a fixed E_x) does not seriously affect the traveling time, which decreases monotonically with increasing E_x.[12] Then the simple picture of streaming motion well explains result (A), whereas it is quite insufficient to explain result (B). We are forced to consider an additional mechanism which gives rise to the abrupt increase in Q_z. Here we should note that in the range $cE_x/H_z < V_{LO}$ the center of the cyclotron motion, $C \equiv (0, -cE_x/H_z, 0)$, enters the surface $|\vec{v}| = V_{LO}$ and accordingly there appears a new area K within the surface, in which electron trajectories never cross the surface $|\vec{v}| = V_{LO}$.[8] The area K forms a spindle-shape region along the z direction bounded by the surface $|\vec{v}| = V_{LO}$ with the center at the point C as described in Fig. 2(b).[1] An electron in the area K, if any, may contribute to Q_z much more efficiently than a streaming electron, since the mean free time of an electron in the area K is believed much longer than that of a streaming electron.[13] Therefore, if we assume (as theoretically predicted by Maeda and Kurosawa[1]) that a certain amount of electrons are accumulated in the area K, the response current Q_z may suddenly increase as H_z increases to cover the range $cE_x/H_z < V_{LO}$ introducing an emergence of the area K. This is just the ob-

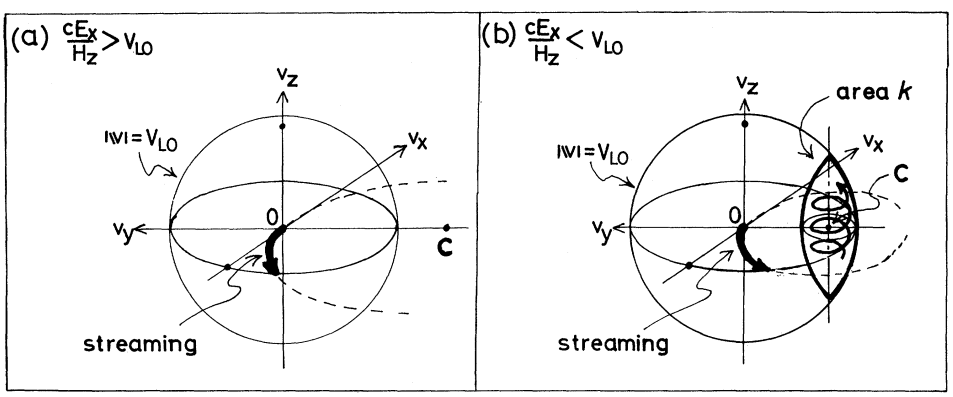

FIG. 2. Anomalous distribution of hot electrons in velocity space for different two conditions of the relative strength of crossed fields $\vec{E} = (E_x, 0, 0)$ and $\vec{H} = (0, 0, H_z)$. The point C denotes the center of the cyclotron orbit, $C = (0, -cE_x/H_z, 0)$. (a) In the range $cE_x/H_z > V_{LO}$, the point C is located outside of the surface $|\vec{v}| = V_{LO}$. The trajectory of a streaming electron is curved by the Lorentz force resulting in an arc around the point C. All the electrons are believed to distribute on the trajectory of streaming. (b) In the range $cE_x/H_z < V_{LO}$, the point C is located within the surface $|\vec{v}| = V_{LO}$. No serious change is involved in the kinetics of streaming electrons, but, in addition to the trajectory of streaming, there emerges an area K in which a certain amount of electrons are accumulated. An electron in the area K is able to produce a large response current Q_z when a perturbing E_z is applied, by drifting along the z direction in a helical motion.

served phenomenon. Thus these results definitely support the interpretation that a population inversion of hot electrons such as described in Fig. 2(b) has been realized in silver halides at 4.2 K in the range $cE_x/H_z < V_{\rm LO}$. Naturally there must exist a mechanism to supply electrons into the area K which leads to the electron accumulation. Kurosawa suggested the following process for the accumulation, on the basis of a Monte Carlo simulation.[14] A streaming electron may happen to overrun the surface $|\vec{v}| = V_{\rm LO}$ to reach a higher-energy state $\epsilon = \hbar\omega_{\rm LO} + \Delta\epsilon$, and to jump into the area K with an excess energy $\Delta\epsilon$ after the LO-phonon emission. Although the probability of such a process may be low, we can believe that the electron accumulation can be realized via this process in the present experiment since (1) the streaming electron very frequently repeats the LO-phonon emission and (2) the electron once jumping into the area K stays in this area for a long time. This type of electron accumulation was first found experimentally in the Hall-effect measurements on silver halides.[2] The present report establishes the interpretation presented in Ref. 2.

We conclude that a definite evidence for the advent of *the population inversion of hot electrons* has been obtained by observing a large helical current within the accumulation area K. More detailed nature of the phenomena, such as the microscopic mechanism of the advent of population inversion, the ratio of population inversion, and scattering mechanism for electrons in the area K, will be discussed in a future paper[15] based on a quantitative analysis of the experimental results.

We are indebted to Dr. F. Moser and Mr. A. Muramatsu for the crystals used. We would like to express our appreciation to Professor T. Kurosawa for helpful discussions. This work was supported in part by the Grants-in-Aid for Scientific Research (A) and for Special Project Research, both from the Ministry of Education, Science and Culture.

[1] H. Maeda and T. Kurosawa, in *Proceedings of the Eleventh International Conference on the Physics of Semiconductors, Warsaw, 1972* (Elsevier, New York, 1972), p. 602.

[2] S. Komiyama, T. Masumi, and K. Kajita, in *Proceedings of the Thirteenth International Conference on the Physics of Semiconductors, Roma, 1976* (Tipografia Marves, Roma, 1976), p. 1222.

[3] K. Kobayashi and F. C. Brown, Phys. Rev. 113, 507 (1959).

[4] H. H. Tippins, Ph.D. thesis, University of Illinois, 1962 (unpublished).

[5] J. W. Hodby, Solid State Commun. 7, 811 (1969); H. Tamura and T. Masumi, J. Phys. Soc. Jpn. 30, 897 (1971).

[6] R. P. Lowndes, Phys. Lett. 21, 26 (1966).

[7] W. Shockley, Bell Syst. Tech. J. 30, 1009 (1951).

[8] I. I. Vosilyus and I. B. Levinson, Zh. Eksp. Teor. Fiz. 50, 1660 (1966), and 52, 1013 (1967) [Sov. Phys. JETP 23, 1104 (1966), and 25, 672 (1967)].

[9] S. Komiyama and T. Masumi, Solid State Commun. 26, 381 (1978).

[10] S. Komiyama, T. Masumi, and T. Kurosawa, in Proceedings of the Fourteenth International Conference on the Physics of Semiconductors, Edinburgh, 1978 (to be published).

[11] S. Komiyama and T. Masumi, in Proceedings of the International Conference on Solids and Plasma in High Magnetic Fields, Cambridge, 1978 (to be published).

[12] The primary interest here exists in the range $cE_x/H_z > V_{\rm LO}/2$. In the range $cE_z/H_z < V_{\rm LO}/2$, the streaming motion becomes impossible (see Refs. 2, 8, and 11).

[13] Collision process of electrons in the area K is believed to be dominated by impurity scattering. The value of $\omega_c\tau_{\rm imp}$ in AgCl MlCB is estimated to reach as high as 12 at 58 kOe.

[14] T. Kurosawa, private communication.

[15] S. Komiyama, K. Kajita, and T. Masumi, to be published.