

Photoinduced electrical domains in ruby

M. I. D'yakonov

A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR

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An explanation is proposed for the appearance of electrical domains in ruby crystals under the action of intense laser irradiation.

It was established in Ref. 1 that an internal electric field, which saturates at a level $E_s \sim 10^6$ V/cm, is produced in concentrated ruby crystals under the action of intense laser irradiation. After the irradiation is switched off, this field remains indefinitely at room temperature, which is explained by the negligible dark electrical conductivity of ruby. The authors interpreted this phenomenon as a photovoltaic effect, arising due to a nonuniform distribution of chromium atoms over polar positions A and B . As a result, the crystal is lacking an inversion center.

It was recently discovered² that irradiation actually produces domains with oppositely oriented (along or against the C_3 axis) electric fields of equal magnitude. Under irradiation in an external field $E_0 < E_s$, the domain structure remains and the field in the domains remains equal to $\pm E_s$, but the total volume of domains with the field oriented along the applied field increases on account of domains with an oppositely oriented field. The domain structure disappears at $E_0 > E_s$, along with any memory of irradiation. This effect depends critically on the chromium concentration^{1,2} and on the temperature under which the specimen is irradiated²: at $T > T_c \approx 140$ K, the domain structure is not formed.

The experimental data listed above are yet to be explained. The interpretation given in Ref. 1 is not consistent with the results in Ref. 2, in particular, with the high uniformity of the field in domains and the existence of a critical temperature.

A qualitative theory of the effect is proposed in this paper. In contrast to Ref. 1, we assume that chromium atoms are distributed uniformly over A and B positions, so that a photovoltaic current cannot appear in the specimen in the absence of an electric field. This current, however, can appear in the presence of a field. The theory is based on the proposition that the photovoltaic current $J(E)$ is oriented *opposite* to the direction of the field. This assumption is consistent with the general principles: It is well known³ that the sign of the photovoltaic current can change with changing wavelength of the light. If the absolute magnitude of $J(E)$ for weak fields exceeds the conduction current, then the state with $E = 0$ will be stable and an electric field will appear spontaneously in the crystal.

The current density j is given by the expression

$$j = \sigma E + J(E), \quad (1)$$

where σ is the conductivity arising under irradiation, and E is the projection of the

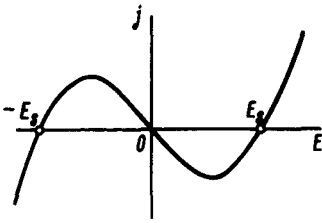


FIG. 1. Suggested dependence $j(E)$. At $j=0$, the states with $E = \pm E_s$ are stable.

field along the C_3 axis. We are examining the one-dimensional case, and the surface of the specimen is perpendicular to the C_3 axis. In order to examine nonuniform distributions of the field, a diffusion current proportional to $-d^2E/dx^2$ must be added to the right side of (1).

For low fields, according to the assumption made, $J(E) = -\alpha E$, where α is a positive coefficient which depends on the intensity and wavelength of radiation, chromium concentration, and temperature. The behavior of the function $J(E)$ for high fields is not important. It can saturate or decrease in absolute magnitude or even change sign. The function $j(E)$ for $\alpha > \sigma$ is shown schematically in Fig. 1. For $j=0$, there is an unstable stationary state with $E=0$ and there are stable states with $E = \pm E_s$. Thus a uniform field, whose sign is determined by the initial fluctuation or by the nature of the inhomogeneities in the specimen, should appear in a disconnected specimen under the action of irradiation.

To understand the experimental results,² it is important to keep in mind that in the absence of an applied voltage the specimen is actually short-circuited, because the time required for charge to flow away from the surfaces of the specimen is much shorter than the characteristic time for development of an instability. A zero voltage is thus maintained on the specimen and a uniform field cannot arise. Under these conditions, the instability of the state with $E=0$ must lead to the formation of two domains with equal volume with fields $\pm E_s$ (Fig. 2a). The width of the domain wall is of the order of the Debye radius, determined by the density of photoexcited carriers.

If during irradiation a voltage $V = -E_0 l$ (l is the width of the specimen) is applied to the specimen and $E_0 < E_s$, then it can be shown that, as before, $j=0$ in the stationary state. In this case, the domain wall will move in such a way that relation $(l_+ - l_-)E_s = E_0 l$ will be satisfied [l_{\pm} are the dimensions of the domains with fields $\pm E_s$ (Fig. 2b)]. The charge density on the domain wall remains after irradiation ceases. If the specimen is disconnected from the voltage source after irradiation ceases, then the charge which comes from the external medium equalizes the potentials of the surfaces of the specimen. In this case, the distribution of the field changes as shown in Fig. 2c.¹⁾ For $E_0 > E_s$, it is impossible for a stationary state with zero current j to arise. In this case, the field in the specimen is uniform (Fig. 2d) and $j = \sigma E_0 + J(E_0)$. Charges producing the field E_0 are now situated only on the surfaces of the specimen. After irradiation ceases and the voltage is removed, they are neutralized and the field in the specimen disappears.

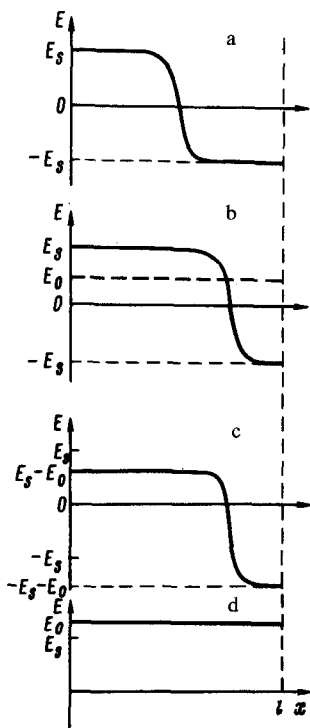


FIG. 2. Electric field distribution in the specimen. a) Zero voltage on the specimen; b) with voltage applied, average field $E_0 < E_s$; c) after irradiation under conditions b, laser beam switched on, and voltage switched off; d) for $E_0 > E_s$.

These conclusions completely agree with the experimental results.² The theory predicts that there is no current in the irradiated specimen for $E_0 < E_s$ (see experimental results in Ref. 4).

We shall now examine one of the possible mechanisms for the appearance of a photovoltaic current in an electric field. Mobile carriers are apparently formed under irradiation in two stages,¹ the first of which consists of populating the metastable 2E level of chromium. The photovoltaic current is evidently proportional to the difference in concentrations of the A^* and B^* chromium ions in positions A and B , excited in the 2E state: $J = k(A^* - B^*)$, where k is a coefficient proportional to the intensity of irradiation. In the presence of a field E , the energies of the metastable levels of ions in positions A and B differ by an amount $2Ed$, where $\pm d$ are the dipole moments of the 2E states for A and B ions. Because of transfer of excitation between A and B ions, there is a tendency for a Boltzmann distribution to form in the metastable states, leading to unequal densities A^* and B^* and thereby to the appearance of a photovoltaic current. In the stationary state, we have

$$A^* - B^* = N \frac{\tau}{\tau + \tau^*(E)} \operatorname{th} \left(\frac{Ed}{T} \right), \quad (2)$$

where N is the total density of chromium ions, which are excited to the 2E state; τ is the lifetime of the state; $\tau^*(E)$ is the time it takes to transfer excitation from ion A to ion B ; this time apparently increases appreciably with the energy gap $2Ed$, as well as with a decrease in the chromium concentration; and T is the temperature in energy units.

We can now write an expression for the photovoltaic current in the form (in accordance with the experimental conditions, we assume that $|Ed| \ll T$):

$$J(E) = -\sigma E \frac{T_c}{T} \frac{\tau + \tau^*(0)}{\tau + \tau^*(E)}, \quad (3)$$

where the critical temperature is $T_c = (-kdN/\sigma)(1 + \tau^*(0)/\tau)^{-1}$. The photovoltaic current is oriented opposite to the field if $kd < 0$. The saturation field E_s is given by the equation, which follows from the condition $j = 0$:

$$\frac{\tau + \tau^*(0)}{\tau + \tau^*(E_s)} = \frac{T}{T_c}. \quad (4)$$

The dependence $E_s(T)$ behaves in a manner characteristic of a second-order phase transition at $T = T_c$.

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¹Because of the inhomogeneity of the specimen, there can conceivably be several domains, rather than two domains. In this case, l_{\perp} should be understood to mean the total length of domains with fields $\pm E_s$.

¹P. E. Liao, A. M. Glass, and L. M. Humphrey, Phys. Rev. B **22**, 2276 (1980).

²S. A. Basun, A. A. Kaplyanskiĭ, and S. P. Feofilov, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 492 (1983) [JETP Lett. **37**, 586 (1983)].

³W. T. H. Koch, R. Munser, W. Ruppel, and P. Wurfel, Solid State Comm. **4**, 915 (1975).

⁴S. A. Basun, A. A. Kaplyanskiĭ, S. P. Feofilov, and A. S. Furman, Pis'ma Zh. Eksp. Teor. Fiz., this issue.

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