Nonresonant microwave absorption in $LiNbO_3$:Mg:Cr(Fe) in low magnetic fields

I. M. Zaritskiĭ, L. G. Rakitina, A. A. Konchits
Institute of Semiconductors, Academy of Sciences of the Ukraine, 252650, Kiev

G. Corradi and K. Polgar
Research Laboratory of Crystal Physics, Academy of Sciences of Hungary, N-1112, Budapest, Hungary

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Nonresonant microwave absorption has been observed in $LiNbO_3$:Mg:Cr(Fe) crystals at $T \leqslant 280$ K in low magnetic fields. This is the first such observation. The effect might be a consequence of a superconductivity of internal regions in crystals which are basically insulators.

Crystals of $LiNbO_3$(LNO) doped with Mg("LMNO crystals") undergo changes in physical properties above a certain threshold Mg concentration. This threshold concentration is 4–6 mole %, depending on the Li/Nb ratio and on whether other impurities are present. The thermal stability and the stability with respect to radiation and laser light increase substantially when these changes occur.[1–3] The ability to retain holographic information disappears.[4] Changes occur in the optical and acoustic properties. The composition of the electron and hole radiation defects is altered in a qualitative way.[5] The ESR spectra of impurity Fe and Cr change radically.[6,7] In this letter we are reporting the first observation of yet another "threshold" property of LMNO: Fe(Cr) crystals: an indication of a superconductivity at $T \leqslant 280$ K. This indication consists of the appearance of a nonresonant microwave-absorption signal near a zero magnetic field, which is detected by an extremely sensitive ESR technique.

Single crystals of $LiNbO_3$ doped simultaneously with Mg (0–8 mole %) and either Fe or Cr were grown by the Czochralski method in the Research Laboratory of Crystal Physics, Academy of Sciences of Hungary. The samples were oriented and cut parallel to the principal axes of the crystal, x, y, z ($z \| \vec{c}$). For the ESR studies we used a Radiopan SE/X-2544 3-cm-range rf spectrometer, with special apparatus for carrying out measurements at very low magnetic fields over the broad temperature range $T = 12$–400 K. Some of the measurements were carried out with a superheterodyne rf spectrometer (which also operated in the 3-cm range). In particular, the latter spectrometer was used for measurements in the configuration $\vec{B} \| \vec{B}$, where $\vec{B}_1$ and $\vec{B}$ are the magnetic component of the microwave field of the cavity and the external magnetic field, respectively.

Figure 1 shows the temperature dependence of the derivative of the microwave-absorption signal, $\partial P / \partial H$, near $\vec{B} = 0$ in LMNO:Cr. We see that the signal appears near $T = 280$ K and increases with decreasing T. Figure 2 shows the temperature dependence of the peak intensity of this signal. Similar results were found for

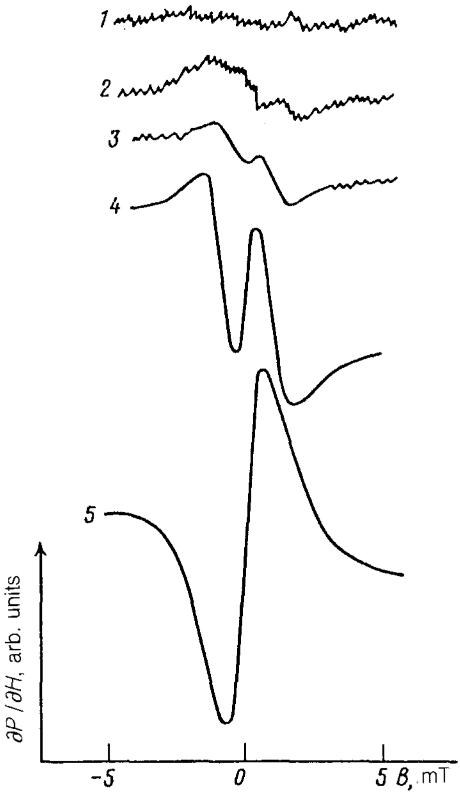

FIG. 1. Derivative of the microwave absorption near $\vec{B}=0$ at various temperatures in LMNO:Cr. T,K: 1—300; 2—280; 3—170 ($A/3$); 4—70 ($A/60$); 5—30 ($A/120$). Here A is the gain. $\vec{B}\,||\vec{c}$.

LMNO:Fe crystals. The shape of these signals and their temperature dependence are qualitatively the same in LMNO:Cr and LMNO:Fe. There is the difference that the LMNO:Fe signals are stretched out more than the LMNO:Cr signals, and the transition from a two-component signal to a one-component signal occurs at a different temperature. Over a broad T range the temperature dependence of the signal amplitude is $I\propto T^n$ with $n\simeq 3.6$–3.7 (Fig. 2). At $T=77$ K we find that the shape of the signal has an angular dependence. In the configuration $\vec{B}\,||\vec{c}$ the signal is a two-component signal (Fig. 1), while with $\vec{B}\perp\vec{c}$ it becomes a single-component signal, whose shape is the same as that of the low-temperature signal (Fig. 1).

We made a special point of determining whether the microwave absorption was resonant or nonresonant. For this purpose we carried out two mutually complementary experiments: We compared the signal intensities in the cases $\vec{B}_1\perp\vec{B}$ and $\vec{B}_1\,||\vec{B}$, and we detected the signal at two microwave frequencies, about 0.5 GHz apart. We found that the intensity of the signal was essentially independent of the relative orientation of $\vec{B}_1$ and $\vec{B}$, and the signal did not shift upon a change in the observation frequency. The ESR line, on the other hand, shifted the expected amount, $\Delta H=(h/g\beta)\Delta\nu$, along the magnetic field (h is Planck's constant, g is the g-factor, and β is the Bohr magneton).

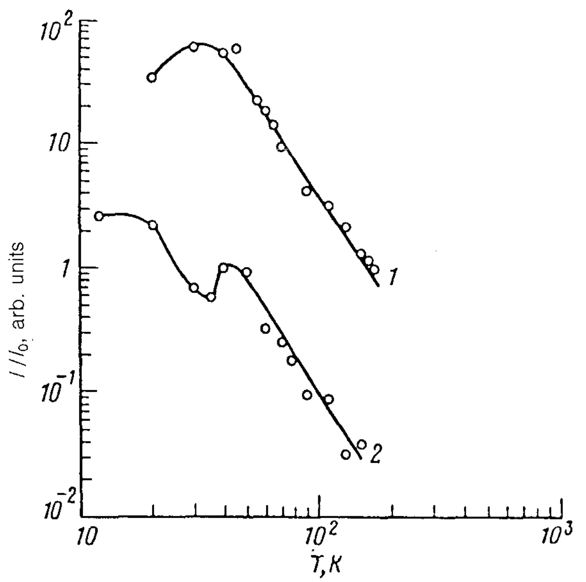

FIG. 2. Temperature dependence of the peak intensity I of the $\partial P/\partial H$ signal near $\vec{B}=0$. 1—LMNO:Cr; 2—LMNO:Fe. Here $I_0 \propto T^{-1}$ (I_0 is the peak intensity of the ESR signals of Cr^{3+} and Fe^{3+} for $1/2 \leftrightarrow -1/2$ transitions).

We thus concluded that the low-field absorption signal is nonresonant. We also found that this signal appears after a threshold is reached, at $C_{Mg} \geqslant 4$ mole % in LMNO:Cr and at $C_{Mg} \geqslant 6$ mole % in LMNO:Fe—at the same time that the modified ESR spectra of Fe^{3+} and Cr^{3+} appear.[6,7]

In the LMNO crystals without other dopants, and also in LMNO:Ti and LMNO:Ni, we found no microwave absorption signal near $\vec{B}=0$. It can thus be concluded that the Fe and Cr dopants along with Mg play a decisive role in the appearance of the signal.

Microwave absorption signals which are nonresonant but which do depend on the magnetic field were discovered in high-T_c superconductors at essentially the same time these superconductors themselves were discovered.[8] These signals are related to the Josephson nature of the superconducting medium. They are a characteristic feature of the transition of these materials to the superconducting state at $T \leqslant T_c^{\text{ons}}$, where T_c^{ons} is the onset temperature of the transition. The unique sensitivity of the ESR technique makes it capable of detecting these signals even when the superconductivity is an "island" superconductivity (as it is, for example, in polycrystalline high-T_c films) and is not detected in transport measurements.[9] The detailed agreement of (a) the shape of the low-field absorption signal in LMNO:Cr(Fe) and (b) the temperature dependence of this signal with the corresponding properties of the high-T_c superconductors[10] lead us to make the serious suggestion that we are dealing with the nucleation of a superconductivity in LMNO:Cr(Fe) at $T \leqslant 280$ K. [A power-law temperature dependence of the signal amplitude is totally uncharacteristic of ESR signals, while it is characteristic of superconductors, where the signal amplitude usually varies as $(T/T_c)^n$.]

Looking at the low-field signal (Fig. 1) from this point of view, we could say that it is of a "thin-film nature"[11] and does not exhibit hysteresis down to $T \simeq 12$ K. These results indicate a low "critical current" and a strong dependence of this critical current on $\vec{B}$ in the superconducting regions. The properties of this signal are similar to those which have been studied in high-T_c-like superconducting superlattices based on PbTe–PbS (Ref. 11). Analysis of the shape of the low-field signal yields an estimate of the average areas of the superconducting regions $(\vec{S})$ and their standard deviation $\Delta\vec{S}$ (Ref. 10):

$$\bar{S} \simeq \frac{\Phi_0}{2\pi 2H_m}; \quad \Delta\bar{S} \simeq 1.15\frac{\Phi_0}{4\pi 2H_m}. \tag{1}$$

Here $\Phi_0 = 2\times 10^{-7}$ G·cm^2 is the flux quantum, and H_m is the value of the magnetic field at the extrema of the low-field signal (Fig. 1). Using the values of H_m (which are approximately the same for Cr^{3+} and Fe^{3+}), we find $\overline{S} \simeq 6\times 10^{-2}$ μm^2 and $\Delta\overline{S} \simeq 0.1$ μm^2. In other words, the average linear dimensions of the microscopic superconducting regions are about 0.25×0.25 μm. Interestingly, a similar situation was found in observations of microscopic superconducting regions in GaAs, which remained a high-resistance material.[9]

Another possible reason for the appearance of a microwave absorption signal near $\vec{B} = 0$ might be the formation of slightly ferromagnetic or spin-glass regions in a paramagnetic matrix.[12] Characteristic features of such a signal are (a) that there is a hysteresis in the absorption over the entire observation temperature range, with the sign opposite that of the hysteresis in the high-T_c superconductors, and (b) that a plot of a shape of the derivative of the absorption has a shape similar to a hysteresis loop. We see from Fig. 1 that these features are not characteristic of the absorption signals which we have observed. It can thus be suggested that an origin of these signals in magnetism is less probable than an origin in the formation of superconducting regions inside the $LiNbO_3$:Mg:Cr(Fe) crystals, which remain insulators on the whole.

Further research will be required to finally resolve the nature of this effect.

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