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Ultrasonic velocities in $(Ti_{1-x}V_x)_2O_3$ in high magnetic fields

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Measurements of the velocities of ultrasonic waves in $(Ti_{1-x}V_x)_2O_3$ crystals with $x \leq 0.09$ have been made as a function of magnetic induction up to 130 kG at liquid-helium temperatures. The results indicate that screening of interionic forces by 3*d* electrons is important in metallic $(Ti_{1-x}V_x)_2O_3$ samples but not in semiconducting Ti_2O_3. This implies that a complete explanation of the semiconductor-metal transition in Ti_2O_3 will need to include the effect of screening.

I. INTRODUCTION

Alloying V_2O_3 into Ti_2O_3 has been found[1] to reduce the temperature dependence of the electrical resistivity in the temperature ranges where Ti_2O_3 is semiconducting (below 400 K) and where it undergoes a transition to metallic behavior (400 to 500 K). In fact a few percent of V_2O_3 is sufficient to cause metallic behavior at most or all temperatures depending on the particular concentration of V_2O_3 present. The influence of V_2O_3 at ordinary temperatures (above 100 K) has been ascribed[1] to the overlapping of the conduction and valence bands comprised mainly of a_{1g}-like and e_g-like Ti-3*d* electronic states, respectively. However, the excess heat capacity[2] observed for $(Ti_{1-x}V_x)_2O_3$ at low temperatures has been explained in other ways. They included invoking a high density of electronic energy states near the Fermi level due to a one-dimensional-like band[2] or to a vanadium impurity band.[3] Another explanation employed conduction-band polarons[4] whose effective mass depended on the screening which the (lighter) holes provided for the Coulomb field around each conduction electron. This polaron model was also used to explain the semiconductor-to-metal transition in Ti_2O_3.

The one-dimensional band model was objected to[5] because of the arrangement of the metal ions in the α-corundum structure. Van Zandt[3] devised a band model which contained vanadium impurity levels each of which was downshifted relative to the respective Ti 3*d* bands derived from a similar type of atomic orbitals. A high density of states occurred at the Fermi level due to a narrow band of e_g-like vanadium orbitals being superimposed on the Ti-3*d* valence band (composed mainly of a_{1g}-like orbitals) near the top of the latter. Recent magnetic-susceptibility data[6] have revealed inadequacies in the Van Zandt model and have been used along with electrical-resistivity results in the construction of still another band model[7] whose description we shall defer until work which preceded it has been discussed.

Support for the presence of two sets of electronic states near the Fermi energy in metallic $(Ti_{1-x}V_x)_2O_3$ has been provided by ultrasonic velocity measurements[8] at liquid-He temperatures. In fact it was inferred that four of the elastic constants depended on strain-induced transfer of electrons between states describable by the Van Zandt model. The ultrasonic data also revealed that all the elastic constants exhibited changes proportional to the change in the *c*-axis lattice parameter caused by alloying with V_2O_3. Lattice-parameter-correlated (LPC) changes in some of the elastic constants (c_{11}, c_{12}, and c_{33}) were interpreted in terms of anionic separation by a consideration of the pressure dependences of the elastic constants of α-Al_2O_3.[9] However, the LPC changes in the other elastic constants (c_{13}, c_{14}, and c_{44}) of $(Ti_{1-x}V_x)_2O_3$ could not be so interpreted. This suggested that the almost close-packed spherical oxygen ion model used for α-Al_2O_3 was not suitable for $(Ti_{1-x}V_x)_2O_3$; instead a more realistic ionic arrangement and the nature of the electronic bonds between ions needed to be considered.[8]

In the hope of obtaining more direct evidence for the role played by 3*d* electrons in the elastic constants and the semiconductor-metal transition in the $(Ti_{1-x}V_x)_2O_3$ system, we began[10] an investigation of the effect of a large magnetic field on the velocities of ultrasonic waves at low temperatures in samples with $0 \leq x \leq 0.09$. The results of our investigation will be reported herein. Before doing so, we shall summarize recent magnetic susceptibility[6,11] and electrical measurements,[7] on $(Ti_{1-x}V_x)_2O_3$ alloys at low temperatures and the band model[7] derived from them.

The magnetic susceptibility of each sample with $0.007 < x < 0.10$ has a Curie-Weiss-like behavior[6] at temperatures approaching the liquid-He region and exhibits spin glass behavior[11] at lower temperatures (i.e., a cusplike maximum in the susceptibility and magnetic remanence effects). The paramagnetism of these, and of lower vanadium content samples, was interpreted as due to localized moments on vanadium ions. Since the experimental paramagnetic susceptibility for $x > 0.02$ samples is greater than that calculated[12] by the Van Zandt model, it has been suggested that much, or most, of the excess heat capacity in metallic $(Ti_{1-x}V_x)_2O_3$ is of magnetic origin.[7,11] Recent electrical resistivity measurements indicate that conduction via localized impurity states is important at low temperatures in undoped Ti_2O_3 and in Ti_2O_3 very lightly doped with V_2O_3.[7] These results have been employed along with the magnetic susceptibility results to deduce the following model[7] for the electronic energy states in $(Ti_{1-x}V_x)_2O_3$: substitution of vanadium for Ti causes there to be Ti^{4+} and V^{2+} levels. At low vanadium concentrations $0 < x < 0.005$, the Ti^{4+} levels lie above the top of the valence band (comprised of Ti $3d$ states). Each V^{2+} ion has three electrons of parallel spin in an energy level below the Fermi energy, which is near the top of the valence band. For $0.01 < x < 0.03$ the Ti^{4+} levels form a partially filled band which merges into the top of the valence band. The bottom of the conduction band lies above this combination band and, at zero temperature, there are fully occupied singlet vanadium levels below the Fermi energy E_F and empty V levels of opposite spin above E_F. Finally for $0.03 < x < 0.10$ the bottom of the conduction band (comprised of e_g-like Ti $3d$ states) merges into the top of the combination band. There is no longer any separation between valence and conduction bands. Again at $T = 0$ K there are fully occupied singlet V states below E_F and empty V states of opposite spin above E_F. This model of the electronic energy levels is able to interpret various electronic properties of $(Ti_{1-x}V_x)_2O_3$ at least qualitatively. However, it does not provide any direct information about the electron-lattice or electron-phonon interactions. These are expected to be important because of Raman,[13] ultrasonic attenuation,[14] piezoresistance,[15] and infrared reflectance[16] results on Ti_2O_3 or Ti_2O_3-V_2O_3 alloys, as well as because of some of the work[1] mentioned previously. In fact theoretical work on Ti_2O_3 has included a free energy treatment[17] connecting lattice distortion, electronic properties, and phonon behavior with only limited success. There has been a band-structure calculation[18] for Ti_2O_3 but it fails to produce a semiconducting gap at low temperatures thereby implying that some additional interaction needed to be considered. The authors of the band calculation suggest Coulombic interactions between ions.

We shall present results which indicate that mobile charge carriers (presumably in $3d$ electronic states associated with the metal ions) play an important role in screening interionic interactions. It turns out that screening influences some of the elastic constants (c_{11} and c_{33}) more than does the strain-induced transfer of electrons between different sets of $3d$ states.[8]

II. EXPERIMENTAL DETAILS

Our present experiment consisted of measuring the transit times of pulses of 30-MHz ultrasonic waves in oriented single-crystal samples of Ti_2O_3 and of $(Ti_{1-x}V_x)_2O_3$ with $x = 0.02$, 0.04, and 0.09 as a function of magnetic field at liquid-helium temperatures. The samples were grown by personnel in the Purdue Central Crystal Growth Facility using the Czochralski-Kyropoulous technique. The V_2O_3 contents were determined by wet chemical analysis performed at the Battelle Memorial Institute, Columbus, Ohio. The magnetic field was provided by a 140-G Intermagnetics General Corporation superconducting magnet which was immersed in liquid helium at its boiling point. The field strength was measured with a copper magnetoresistance probe installed in the magnet. The pulse-echo-overlap method[19] was used for transit time measurement , and each sample was prepared and had a quartz transducer bonded to it as described elsewhere.[20] The sample was mounted on a holder immersed in an insert Dewar containing liquid helium whose vapor pressure was controlled and measured to establish the temperature of the sample. The temperature was also monitored by means of a Ge resistance thermometer. However, in view of its magnetoresistance the Ge thermometer was used to deduce an actual value for the temperature only when no magnetic field was present, i.e., when the magnet was in the virgin state.

III. RESULTS

In Fig. 1 are shown the fractional changes in ultrasonic velocities as a function of magnetic induction for a $(Ti_{0.98}V_{0.02})_2O_3$ crystal at 4.2 K. It can be seen that the velocities of longitudinal waves increase quadratically with magnetic induction by an amount which is independent of whether the propagation direction ($\vec{q}$ vector) of the wave is parallel or perpendicular to the magnetic induction ($\vec{B}$), or parallel to the a axis or the c axis of the crystal. The fractional increase is given by $9.3 \times 10^{-9} B^2$(kG). On the other hand, the shear wave velocity is not changed by the magnetic field. The shear wave data are for fast transverse waves whose plane of polarization makes an angle of $\frac{1}{2}\arctan[4C_{14}/(C_{11} - C_{12} - 2C_{44})] \simeq 16.5°$ with the basal plane of this α-corundum structure. The velocity of these shear waves depend on the C_{11}, C_{12}, C_{44}, and C_{14} elastic constants, but because

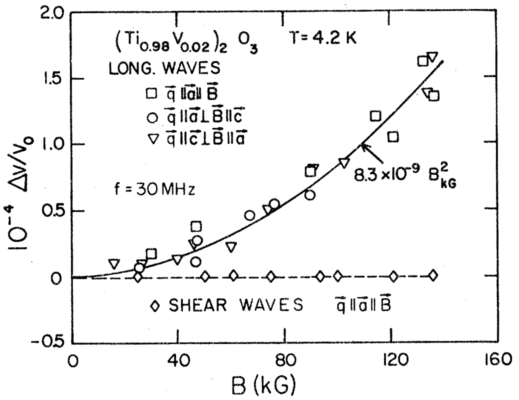

FIG. 1. Fractional change in the velocity of ultrasonic waves *versus* magnetic induction for a metallic $(Ti_{1-x}V_x)_2O_3$ sample with $x = 0.02$; $\vec{q}$ is the wave propagation vector, $\vec{a}$ and $\vec{c}$ are lattice parameter vectors, and $\vec{B}$ is the magnetic induction.

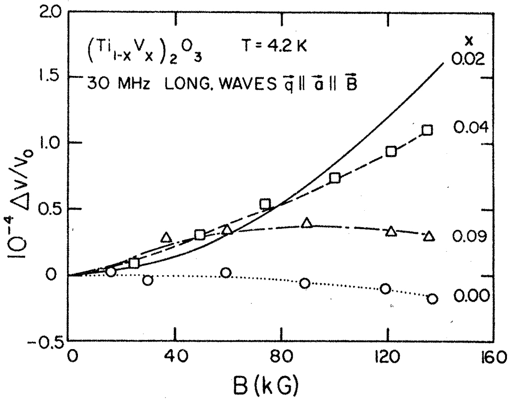

FIG. 2. Fractional change in the velocity of longitudinal ultrasonic waves *versus* magnetic induction in semiconducting Ti_2O_3 ($x = 0.00$) and metallic $(Ti_{1-x}V_x)_2O_3$ ($x = 0.02$, 0.04, and 0.09). The vector symbols have the same meaning as in Fig. 1. Data points have been omitted for the $x = 0.02$ sample to avoid clutter.

of the smallness of C_{14}, is very nearly equal to $[(C_{11} - C_{12} + C_{44})/2\rho]^{1/2}$, where ρ is the density, thus implying that not only this type of shear wave but also those depending on C_{44} or $C_{11} - C_{12}$ alone are not affected by the magnetic field. (Additional types of shear waves were not studied for this sample because of the great difficulty of bonding a shear wave transducer well enough so that it remained intact during the temperature changes occurring as the sample holder was moved into the position in the cold insert Dewar.)

Figure 2 shows data for undoped Ti_2O_3 and for samples with various concentrations of V_2O_3 alloyed in Ti_2O_3. From Fig. 2 it can be seen that in Ti_2O_3 longitudinal waves are not stiffened by the magnetic field. (The very slight decrease in $\Delta v/v_0$ might be due to a slight increase in temperature because of poorer than normal temperature control.) Figure 2 also shows that metallic $(Ti_{1-x}V_x)_2O_3$ samples with $x = 0.04$ and 0.09 have their longitudinal sound velocities increased by a magnetic field. A curve summarizing the data for our $x = 0.02$ sample is also included, but the data points have been omitted since they are shown in Fig. 1. From the results shown in Fig. 2, it appears that the presence of mobile charge carriers is necessary for stiffening to occur, but that the form as well as the amount of stiffening depends on the vanadium concentration. Thus as already noted for $x = 0.02$, the fractional increase in velocity $\Delta v/v_0$ is quadratic in magnetic field strength. However, for $x = 0.04$, $\Delta v/v_0$ is roughly proportional to $B^{1.4}$, and lies above the $x = 0.02$ curve at the lower fields and below the $x = 0.02$ curve at the highest fields. Finally, for $x = 0.09$, $\Delta v/v_0$ increases about as strongly at low fields as it did for $x = 0.04$, but saturates at higher fields. For metallic samples at the highest fields $\Delta v/v_0$ is less the larger the V_2O_3 content.

We do not have any shear wave velocity versus magnetic field data to report on samples with $x = 0$, 0.04, or 0.09. We attempted to obtain such data but were unsuccessful due to bond failure. Since even our $x = 0.02$ sample, which showed the largest longitudinal wave stiffening, exhibited no measurable change in shear wave velocity due to the magnetic field, it could hardly be expected that there would be any field dependence of shear wave velocities in our other samples. Thus, we did not feel justified in continuing efforts to obtain shear wave data on the latter.

In order to determine the effect of temperature on the magnetic stiffening of longitudinal waves we made measurements for an $x = 0.02$ sample at 1.4 K and for an $x = 0.04$ sample at 2.0 K. It was found that the $x = 0.02$ sample exhibited stiffening at 1.4 K identical to that at 4.2 K, and that the $x = 0.04$ sample exhibited a stiffening at 2.0 K which was larger than that at 4.2 K by an amount equal to the combined experimental errors. This lack of significant temperature dependence justifies the zero-temperature assumption we shall use in analyzing our data.

IV. DISCUSSION

Certain features of our results rule out their being due to effects which might otherwise be thought important. Thus the lack of stiffening of shear waves provides evidence that magnetostriction is not important in determining the ultrasonic transit times we measure. In addition the longitudinal wave stiffening is more than an order of magnitude larger than our estimates of the magnetostriction.[21]

Stiffening of longitudinal waves, but not of transverse waves, propagating along the magnetic field direction also indicates that deflection of moving charges (lattice ions and electrons responding to the ultrasonic waves) by the magnetic field cannot be responsible for the effects we observe. If it were, the shear waves and only longitudinal waves propagating perpendicular to the magnetic field would have been stiffened as has been found in metals.[22]

The size of the longitudinal wave stiffening which we observed is much too large to be explainable in terms of interband electron transfer in view of the small contribution this mechanism seems to make to the elastic constants of $(Ti_{1-x}V_x)_2O_3$.[8] This is in contrast to the case of V_3Si in which the stiffening of longitudinal ultrasonic waves traveling along the magnetic field direction has been attributed to the influence of the magnetic field on interband electron transfer.[23]

We now turn to the mechansim which we believe is responsible for our experimental findings. It involves the separating of electronic energy bands into subbands of opposite spin due to the presence of the magnetic field. A given spin band is displaced relative to its counterpart with opposite spin so that a redistribution of electrons will occur. This causes a reduction in the screening of interionic forces provided by carriers near the Fermi energy and thus a stiffening of the lattice and an increase in ultrasonic velocity.

Although no theoretical expression is available for velocity change due to a magnetic field affecting d-electron screening, there does exist theoretical work which is useful for interpreting our results as we now indicate. Quite some time ago, a simple calculation[24] was made for the increase in velocity of longitudinal sound waves due to a magnetic field reducing the screening provided by conduction electrons in a metal. The velocity increase was proportional to the square of the magnetic field but independent of the angle between the magnetic field and the direction of sound propagation. These characteristics are found in our results (see Fig. 1) indicating that screening is probably involved. Unfortunately Ref. 24 cannot be expected to provide a very quantitative description of our results since it treated the conduction electrons as a gas. Later work[25] has used a more realistic model which included contributions to the velocity coming from exchange and correlation effects between electrons, band structure effects, and umklapp processes, as well as the electron gas contribution. However, this work did not consider the influence of a magnetic field, and since it was concerned with the alkali metals and aluminum, the issue of screening by d electrons did not arise.

More recently the special character of d-electron screening has been treated theoretically,[26] but only for zero magnetic field. Numerical calculations based on this theory have been made of phonon spectra and elastic constants of some transition metals[27,28] and a transition-metal compound.[28] It was shown[27] that the quasilocalized character of the d electrons causes local-field corrections, contained in nondiagonal elements of the dielectric matrix, which influence longitudinal phonons and some of the elastic constants but, at least for bcc and fcc transition metals, do not affect transverse modes; these transverse modes are affected by s-type electrons, however.

Our observation of magnetic stiffening for longitudinal ultrasonic modes, but not for transverse modes, is thus consistent with the absence of s-type electrons in our samples and the behavior to be expected on the basis of theory for d-electron screening in zero magnetic field. To make a semiquantitative analysis of the data for our $x = 0.02$ sample we shall present some expressions obtained by following Ref. 25. The first shows the square of the velocity as the sum of terms which involve screening, ionic core, and umklapp effects

$$v^2 = v_s^2 + v_c^2 - v_u^2 \,. \qquad (1)$$

Of the terms on the right-hand side of Eq. (1) only v_s^2 is magnetic field dependent so that the change in velocity with field is

$$\frac{\Delta v}{v} = \left(\frac{v_s^2}{v_s^2 + v_c^2 - v_u^2}\right)\left(\frac{v_s(H) - v_s(0)}{v_s}\right). \qquad (2)$$

In order for Eq. (2) to be useful, an evaluation of the first factor is needed. This can be done only approximately especially since we are dealing with a compound rather than with a monatomic metal. By using the monatomic metal model of Ref. 5, but allowing for more than one band we have

$$v_s^2 = \frac{4\pi N Z^2 e^2}{KM} \sum \left(\frac{1}{k_s^2} - \frac{1}{2(k_F^2 + k_s^2)}\right), \qquad (3)$$

where N, Ze, and M are ionic concentration, charge, and mass, respectively, and K is the static dielectric constant. The sum is over partially filled bands. For each band the screening vector k_s is given by[29]

$$k_s^2 = 4\pi e^2 \frac{dn}{dE_F} \Big/ K, \qquad (4)$$

where e is the electronic charge and dn/dE_F is the rate of change of carrier concentration with Fermi energy and k_F is the Fermi wave vector. For our $x=0.02$ sample k_s^2 and k_F^2 will be small since K is probably large, it has a value of 45 for $\mathrm{Ti_2O_3}$,[16] and n is small, being approximately equal to the vanadium concentration.[30] This can be seen by noting that for a simple parabolic band $k_s^2=6\pi ne^2/KE_F=48(\frac{1}{3}\pi)^{2/3}e^2m^*n^{1/3}/Kh^2$ and $k_F^2=(3\pi^2n)^{2/3}$. The smallness of k_s^2 and k_F^2 implies that v_s^2 will be larger relative to the other terms on the right-hand side of Eq. (1) than it is in ordinary metals. Since the first factor in Eq. (2) has a value near 0.4 for all the alkali metals except Li and is about 1.7 for Al,[25] we take it to be unity for our $x=0.02$ sample.

Using the fact that a magnetic field causes a band to split into subbands of opposite spin which are shifted by an amount $2\mu B$ relative to each other, one finds that $\Delta v_s/v_s$ is proportional to $(\mu B/E_F)^2$, where μ is the magnetic moment of the charge carriers having Fermi energy E_F relative to the band edge in zero field. If the band is parabolic,

$$\Delta v/v=\tfrac{1}{8}(\mu B/E_F)^2. \qquad (5)$$

Using the experimental value of $8.3\times10^{-9}\ \mathrm{kG}^{-2}$ indicated in Fig. 1 in Eq. (5), we calculate that $E_F=22.5$ meV and a Fermi temperature of 260 K. For a parabolic band with a concentration of carriers equal to that of vanadium this gives an effective mass of $13.5m_0$ which is much larger than the valence band mass of $5m_0$ obtained for nonstoichiometric $\mathrm{Ti_2O_3}$ from transport measurements.[31] This result is consistent with the presence of a high density of states at the Fermi energy inferred from interpreting the excess heat capacity as an electronic band effect. However, the Fermi temperature we obtain is much larger than found for the high density of states one-dimensional band of Ref. 2 or the vanadium impurity band of Ref. 3. The quantitative discrepancy may be due to some of the specific heat being of magnetic origin as suggested in Refs. 7 and 11. Experimental evidence for a magnetic contribution is provided by the fact that the heat capacity is affected appropriately by a magnetic field.[32]

For our $x=0.04$ and 0.09 samples the stiffening does not have the B^2 dependence expected for carriers in a simple well-behaved band. This is not too surprising in view of the complications in the band structure adduced from previous work. We *do* think that the velocity stiffening in these samples is due to the magnetic field reducing the screening of interionic forces provided by mobile charge carriers. The strange field dependences of the stiffening might be due to the conduction band having merged into the top of the lower-lying band, first considered to be the valence band[1] and later to be a combination valence-impurity band.[3,7] Screening is affected because the Fermi energy is in the region where the conduction band overlaps the lower band. The conduction-band component itself we expect to have a high effective mass because in undoped $\mathrm{Ti_2O_3}$ the conduction band is narrower than the valence band.[33] Furthermore, the conduction-band component is only slightly occupied and the Fermi energy lies only a short distance above its edge. Thus the "effective Fermi energy" to be used in a relation like Eq. (5) is quite small, resulting in a relatively large $\Delta v/v$. This gives a qualitative explanation of why the initial rise in $\Delta v/v$ with B is so strong for our $x=0.04$ and 0.09 samples. The saturation of $\Delta v/v$ for our $x=0.09$ sample can be explained as a consequence of the magnetic field having split and shifted the spin subbands sufficiently so that the uppermost conduction subband is no longer occupied at all. The absence of saturation for the $x=0.04$ sample could be a consequence of there not being enough electrons to give the required occupancy of the conconduction-band states.

When the valence and conduction-band merger or overlap has taken place, the magnetic field might also have an indirect effect on screening of interionic interactions. This indirect effect could arise because the screening of the ions provided by the heavier conduction-band component electrons is dependent on how the lighter valence-band holes screen the Coulomb forces between the electrons.

Up to this point, the explanation we have presented for our experimental results has not included any paramagnetic ion effects even though magnetic susceptibility data[6] indicate the presence of localized interacting magnetic moments in the $(\mathrm{Ti}_{1-x}\mathrm{V}_x)_2\mathrm{O}_3$ system we are dealing with. We have proceeded thus because investigations involving local moments indicate, or imply, behavior for velocity in a magnetic field different than that which we observe. Thus theory[34] for the acoustical modulation of the g factor or of the spin-orbit interaction, predicts a dependence on the crystallographic orientation of the magnetic field and/or a decrease in velocity as B increases. On the other hand, for rare-earth metals[35] in the paramagnetic state an increase in the velocity of c-axis longitudinal waves is produced by a *transverse* magnetic field. Finally, in metallic alloys the longitudinal sound velocity depends on magnetic field strength only below the spin glass temperature, and then it is decreased by the field with the magnitude of the decrease going through a maximum as a function of temperature.[36] It should be noted data on our samples are for temperatures above T_g.

Certain ultrasonic effects which occur in paramagnets have been attributed to screening of phonons by conduction electrons. These effects include the softening of longitudinal phonons in zero field as the Curie temperature is approached from above due to exchange enhancement of the screening[37] and the increase of the ultrasonic attenuation by an applied field in rare-earth metals due to splitting of the conduction spin subband by the exchange field as the magnetic field induces long-range ordering of 4*f* spins.[38] (An increase in attenuation implies a decrease in velocity.) Thus these effects are also unlike what we observe.

The presence of paramagnetic ions in our samples might influence how screening charge is affected by a magnetic field and thereby effect the ultrasonic velocity, especially if the screening charges provide the coupling between the ionic magnetic moments. The behavior of the velocity stiffening in our $x = 0.09$ sample might then be viewed as a consequence of the (ionic) magnetization falling below the proportionality to field which it has at low fields.[39] In view of our data such an effect would seem to be important only if there are sufficiently large densities of paramagnetic ions and appropriate screening carriers.

V. CONCLUSION

In conclusion, we have obtained new information about the semiconductor-metal transition in the $(Ti_{1-x}V_x)_2O_3$ system for $x < 0.1$ by measuring ultrasonic velocities in high magnetic fields at low temperatures. We find that screening of interionic forces by mobile charge carriers is important in metallic $(Ti_{1-x}V_x)_2O_3$ samples with $x \leqslant 0.09$, but not in semiconducting Ti_2O_3. Thus suggests that screening affects are also important for the semiconductor-metal transition which Ti_2O_3 undergoes between 400 and 500 K. Further theoretical work needs to be done before quantitative use can be made of the magnetic stiffening of ultrasound in metallic $(Ti_{1-x}V_x)_2O_3$. Our work suggests the approach future theory should take.

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